ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

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Section I

Isotopes and Radiation Technology

Isotope Production and Development

The U. S. Radioisotope Industry—1966

(Editors' Note: The USAEC Division of Industrial Participation has for several years prepared a yearly summary of the status of the nuclear industry. The report[1] for the year, *The Nuclear Industry—1966*, was released at the annual joint fall meeting of the Atomic Industrial Forum and the American Nuclear Society in Pittsburgh, Pa., Nov. 3, 1966. The radioisotopes section of this report is presented here because it provides a particularly good summary of the current state of the radioisotopes industry. The editors thank Ernest B. Tremmel, Director of the Division of Industrial Participation, for permission to use this section and John N. Maddox of the Division of Isotopes Development and Robert J. Berte of the Division of Industrial Participation, who reviewed and updated the lists of suppliers.)

Abstract: The status of the commercial U. S. radioisotope industry in 1966 is reviewed in four categories: producers, processors, source fabricators, and equipment manufacturers. Among the significant actions taken during the year by both the AEC and industry which should strengthen the industry both now and in the future were the following:

1. The AEC withdrew from the production of 19 more isotopes, bringing to 34 the total number of isotopes that are no longer produced and sold by the AEC.

2. The first production of ^{60}Co in a commercial power reactor was started.

3. The Martin Marietta Corp. announced a fixed-price isotope-powered generator for sale and sold the first unit to the Navy.

It has now been over 20 years (Ref. 2) since the first shipment of radioisotopes from Oak Ridge National Laboratory (ORNL), and the possibilities for growth in the radioisotopes segment of the nuclear industry continue to appear good. In fact, in such major areas as industry and medicine, the period of greatest growth still may well be in the future.

Although in the United States the packaging and distribution of radioisotopes are performed almost entirely by industry, the AEC is still a principal domestic producer of radioisotopes. However, in 1966, industry has continued to add capability and is showing an ever-increasing interest in this field.

During the year, radioisotope-fueled systems for auxiliary power for a variety of uses came into prominence as their value for space missions requiring up to 10 kw became recognized by agencies developing space vehicles. The prospective demand for fuel for these systems stimulated a variety of plans for production or separation of isotopes useful for these purposes. Another program, developmental work on a radioisotope-powered cardiac pacemaker, is under way; and studies indicate that an artificial heart powered with a radioisotopic source could become a reality.

The AEC formally opened an Isotopes Information Center at ORNL in 1966 to collect, evaluate, and disseminate worldwide information on production of radioisotopes and their use in industry and research.

For the purpose of our report, we have divided the radioisotope industry into four categories: producers, processors, source fabricators, and equipment manufacturers.

Producers

A radioisotope producer is defined as one who can irradiate targets in either a cyclotron or a reactor or can recover fission products from high-level reactor wastes. He may also be a processor (see below).

CYCLOTRON RADIOISOTOPES

As a result of work conducted by AEC and industry, a sizable market, estimated at $0.5 to $1 million per year, has been developed for cyclotron-produced radionuclides. Up to now, the United States has been the dominant supplier of such radioisotopes, although the United Kingdom, South Africa, and the Union of Soviet Socialist Republics also compete in the world market for supplying these materials.

In 1955 the AEC discontinued processing and distributing cyclotron-produced isotopes. Thus, except for industry's dependence for irradiation of targets upon the University of Pittsburgh cyclotron,[3] which became available this year, or on the ORNL 86-in. cyclotron,[4] the industry supplies these radioisotopes. Cyclotron products such as ^{22}Na, ^{57}Co, and ^{84}Rb are sold to radiopharmaceutical suppliers for use in medical, industrial, and fundamental research programs. Among the most promising gamma or positron emitters which are not routinely available, but can be produced by cyclotron techniques, are ^{11}C, ^{13}N, ^{15}O, ^{55}Fe, ^{199}Hg, ^{49}Cr, ^{61}Cu, ^{62}Zn, and ^{73}Se.

The growth in cyclotron-produced radioisotopes may be estimated from the AEC revenue for special

cyclotron target irradiations which amounted to ~$126,000 in FY 1964, ~$259,000 in FY 1965, and ~$275,000 in FY 1966. The principal commercial suppliers are:

Abbott Laboratories, North Chicago, Ill.
Cambridge Nuclear Corporation, Cambridge, Mass.
New England Nuclear Corp., Boston, Mass.
Nuclear Science and Engineering Corporation, Pittsburgh, Pa.
U. S. Nuclear Corp. (subsidiary of International Chemical & Nuclear Corp.), Burbank, Calif.
Nuclear Consultants Corp. (Division of Mallinckrodt Chemical Works), St. Louis, Mo.

REACTOR RADIOISOTOPES

The AEC is still a principal domestic producer and distributor of reactor-produced radioisotopes. However, industry continues to show increasing interest in, and capability to produce and distribute, these nuclides. There has been an increasing emphasis on producing higher-specific-activity material, especially for radiopharmaceutical applications.

The following companies offer radioisotope target irradiation services: (1) General Electric Company, Vallecitos Nuclear Center (Pleasanton, Calif.)[5]; (2) Union Carbide Corporation (Tuxedo, N. Y.); (3) The Babcock & Wilcox Co. (Lynchburg, Va.); and (4) Western New York Nuclear Research Center (Buffalo, N. Y.).[6] In the future, however, additional private reactors, such as power reactors, should become available for large-volume and low-specific-activity radioisotope production. A first approach to the use of power reactors for isotope production was announced this year after AEC approved the production of ^{60}Co in the Big Rock Point nuclear power reactor. Consumers Power Company, Charlevoix County, Mich., is the producer, in cooperation with Neutron Products, Inc., subsidiary of Nuclear Utility Services, Inc., of Washington, D. C.[7]

Since 1963 the AEC has withdrawn[8] from routine production of 34 reactor-produced isotopes (Table 1). In March 1965 they adopted, and published in the *Federal Register*, formal policies and procedures for the orderly transfer of commercial AEC radioisotope production and distribution activities to private industry. These guidelines have been followed for the past 2 years.

Table 1 ISOTOPES NO LONGER ROUTINELY PRODUCED BY AEC

Withdrawal year	Isotopes
1961	^{60}Co (with specific activity greater than 30 curies per gram, increased to 45 curies/g in 1965)
1963	^{125}I, ^{131}I
1964	^{141}Ce, ^{58}Co, ^{51}Cr, ^{134}Cs, ^{55}Fe, ^{85}Sr
1965	^{45}Ca, ^{59}Fe, ^{125}Sb, ^{75}Se, ^{113}Sn, ^{65}Zn
1966	^{110m}Ag, ^{76}As, ^{77}As, ^{198}Au, ^{199}Au, ^{82}Br, ^{109}Cd, ^{115}Cd. ^{115m}Cd, ^{64}Cu, ^{197}Hg, ^{203}Hg, ^{42}K, ^{140}La, ^{99}Mo, ^{24}Na, ^{32}P, ^{35}S (sulfate), ^{124}Sb

Industrial interest in the use of radioisotopes has increased steadily in the last few years. Although industry is producing and distributing more radioisotopes each year, AEC sales (distributions outside the AEC) have also increased as shown below, due primarily to the distribution of large quantities of ^{60}Co for irradiators:

Fiscal year	Value
1963	1.2×10^6
1964	1.3×10^6
1965	1.6×10^6
1966	2.0×10^6

Although statistics are not currently available on industrial sales, indications are that they are rapidly increasing.

Effective May 1, 1966, the AEC increased prices for 13 radioisotopes to recover full costs of production and distribution.[9]

On Dec. 18, 1965, the AEC revised their ^{60}Co price schedule to encourage development of large-scale industrial applications and to assist American firms in competing more effectively in the world market for radiation systems.[10] The AEC's Advisory Committee on Isotopes and Radiation Development reviewed this price revision prior to its acceptance by the Commission and agreed that this action would encourage industrial use and stimulate further development of new applications.

In 1961 both Westinghouse and General Electric started producing limited quantities of high-specific-activity ^{60}Co in their test reactors. The Westinghouse Test Reactor was subsequently shut down, and today General Electric is the only commercial firm engaged in the production of substantial quantities of predominantly teletherapy- and radiography-grade high-specific-activity ^{60}Co.

As previously mentioned, Neutron Products, Inc., recently initiated a joint program with Consumers Power Company to produce megacurie quantities of ^{60}Co in the Big Rock Point power reactor. The specific activity of this ^{60}Co will range from 15 to 60 curies/g, and the product will be used primarily in sources for radiation processing. We expect that power-reactor-produced ^{60}Co will gradually serve a portion of the market now being supplied by the Commission as requirements for radiation processing sources continue to grow.

The problem of foreign competition in the ^{60}Co market has been and continues to be a problem for American firms. In addition to price competition, the Canadian and U. K. firms also provide a more complete package of services than is yet available from individual U. S. companies.

Reactor-produced radioisotopes are used in a large number of applications — medicine, agriculture, crime detection, industrial gages, and space, to name only

a few. The future potential dollar market for radioisotopes is difficult to estimate, but it is growing steadily.

FISSION-PRODUCT RADIOISOTOPES

A third source of radioisotopes is AEC high-level reactor wastes from which fission products are separated.[11] The fission-product radioisotopes of principal interest are ^{90}Sr, ^{144}Ce, ^{137}Cs, and ^{147}Pm, which can be made available in large quantities from by-products of commercial chemical processing plants such as Nuclear Fuel Services, Inc., when their production is warranted by market demands. In 1965 the AEC announced sharp price reductions on these four radioisotopes.[12] Thus, the expanding uses of these isotopes may eventually create an additional source of revenue for utilities.

Fission-product radioisotopes are used in a number of industrial applications as well as for heat and power sources. Promethium-147 and ^{90}Sr are used in luminous sources and atomic "batteries." Strontium-90 is also used in industrial thickness gages and for the treatment of certain eye disorders. Cerium-144 is used for various research purposes and has good potential for use as a heat source. Cesium-137 is used as a radiation source for both industry and medicine and competes with ^{60}Co in certain applications. AEC sales of these isotopes are given below:

	FY 1965		FY 1966	
	Activity, curies	Value	Activity, curies	Value
^{137}Cs	350,989	$192,000	70,002	$ 27,000
^{147}Pm	7,174	28,000	7,511	12,000
^{90}Sr	1,015	3,000	109,262	65,000
^{144}Ce	66	Negl.	138	Negl.
Total	359,344	$223,000	186,913	$104,000

Processors

A processor is defined as one who can prepare targets, separate desired radioisotopes from targets, and make radiochemicals or radiopharmaceuticals for ultimate distribution. Industry continues to assume more and more of these activities. The principal commercial processors are listed in Table 2.

Union Carbide Corporation entered the radiopharmaceutical field this year with their recently acquired Neisler Laboratories, Inc. Mallinckrodt Chemical Works acquired Nuclear Consultants Corp. and Nuclear Research Chemicals, Orlando, Fla. The G. D. Searle & Co. and Nuclear-Chicago Corporation merged, thereby giving Nuclear-Chicago the opportunity to expand into nuclear medicine. Isotopes, Inc., has acquired Hazleton Nuclear Science Corp., Volk Radiochemical Co., and U. S. Nuclear Corp., the latter having previously acquired Isotopes Specialties, Inc. International Chemical & Nuclear Corp.

Table 2 PRINCIPAL PROCESSORS OF RADIOCHEMICALS AND RADIOPHARMACEUTICALS

	Radiochemicals	Radiopharmaceuticals
Abbott Laboratories, North Chicago, Ill.	X	X
Atomic Corporation of America, San Fernando, Calif.	X	
Bio-Rad Laboratories, Richmond, Calif.	X	
Calbiochem, Los Angeles, Calif.	X	
Cambridge Nuclear Corporation, Cambridge, Mass.	X	X
General Nuclear, Inc., Houston, Tex.	X	X
General Radioisotope Processing Co., San Ramon, Calif.	X	
Hastings Radiochemical Works, Houston, Tex.	X	
International Chemical & Nuclear Corp., subsidiary: U. S. Nuclear Corp., Burbank, Calif.	X	
Isotopes, Inc., subsidiaries: Hazleton Nuclear Science Corp., Palo Alto, Calif. Volk Radiochemical Co., Burbank, Calif.	X	
New England Nuclear Corp., Boston, Mass.	X	
Nuclear-Chicago Corporation (Division of G. D. Searle & Co.), Des Plaines, Ill.	X	
Nuclear Consultants Corp. (Division of Mallinckrodt Chemical Works), St. Louis, Mo.	X	X
Nuclear Research Chemicals, Inc., (Division of Mallinckrodt Chemical Works), Orlando, Fla.	X	
Nuclear Science and Engineering Corporation, Pittsburgh, Pa.	X	
Nucleonic Corporation of America, Brooklyn, N. Y.	X	
Schwarz Bio Research, Inc., Orangeburg, N. Y.	X	
E. R. Squibb & Sons, New Brunswick, N. J.		X
Tracerlab (Division of Laboratory for Electronics, Inc.), Waltham, Mass.	X	

purchased Baird-Atomic's radiochemical production facilities and U. S. Nuclear from Isotopes, Inc. Nuclear Science and Engineering Corporation set up a wholely owned subsidiary, Radioactive Materials Corporation, in Buffalo, N. Y., to process and distribute short-lived reactor-produced isotopes.

The estimated total annual sales of radiochemicals is $6 million, and of radiopharmaceuticals, $7 million. Commercial sales of radiopharmaceuticals are increasing 20 to 30% per year.

Source Fabricators

At present some 50 commercial firms are producing alpha, beta, gamma, and neutron sources. Although the alpha- and neutron-source business is small, about $500 thousand per year, and may continue to increase slightly over the years, the beta- and gamma-source business can be expected to increase substantially as a result of increased interest in heat

and radiation processing applications. A number of firms that own large hot cells — Battelle Memorial Institute, Babcock & Wilcox, General Electric, Gamma Process Corporation (through Westinghouse), and Lockheed-Georgia — have bid on large ^{60}Co source encapsulation work.

Nuclear Materials and Equipment Corp. (NUMEC), Apollo, Pa., and Monsanto Research Corporation, Dayton, Ohio, are the principal commercial suppliers of plutonium and americium neutron sources.

Equipment Manufacturers

The six principal types of radioisotopes and irradiator equipment are: radiography units, teletherapy units, gages, self-illuminating devices, isotopic power devices, and irradiators for the radiation processing industry.

RADIOGRAPHY UNITS

Radiography units are manufactured by industry and not by the AEC. The commercial firms active in this field are:

The Budd Company, Phoenixville, Pa.
Gamma Industries, Inc., Baton Rouge, La.
Radionics, Inc., Norristown, Pa.
Picker X-Ray, Technical Operations, Inc., Burlington, Mass.
U. S. Nuclear Corp., Burbank, Calif. (subsidiary of International Chemical & Nuclear Corp.).

The annual volume of sales in this field is approximately $1 million and is expected to increase. The radioisotope units, which use ^{192}Ir, ^{60}Co, ^{137}Cs, ^{170}Tm, and others, compete with machines.

TELETHERAPY UNITS

The eight commercial firms that supply teletherapy units are:

W. F. & John Barnes Company, Automated Machine Division of the Babcock & Wilcox Co., Rockford, Ill.
General Electric Company, Milwaukee, Wis.
Tracerlab (Division of Laboratory for Electronics, Inc.), Waltham, Mass.
North American Philips Company, Inc., New York
Picker X-Ray Corp., Cleveland, Ohio
Schick X-Ray Company, Chicago, Ill.
Siemens New York, Inc., New York
Westinghouse Electric Corporation, Baltimore, Md.

Four of the eight suppliers (General Electric, North American Philips Company, Siemens New York, and Schick X-Ray Company) are distributors of foreign-made units. Of the approximately 1700 teletherapy units in the world, 700 are in North and South America. The annual volume of sales for these units is approximately $2 million. The average new unit being installed in the United States contains 3 to 4 thousand curies of high-specific-activity ^{60}Co with some containing up to 8 to 10 thousand curies.

GAGES

Between 20 and 30 firms are producers of nuclear gages. These gages are used industrially for controlling various processes; measuring flow rates, thicknesses, levels, and densities; and monitoring composition of material by activation analysis. Estimated 1966 sales of industrial nuclear gages are $13 to $16 million, including exports of about $3 million, but excluding rental or lease revenue, which is estimated to be approximately $4 million.

The principal suppliers of industrial nuclear gages and gaging systems are:

The Foxboro Company, Foxboro, Mass.
Industrial Nucleonics Corp., Columbus, Ohio
Nuclear-Chicago Corporation (Division of G. D. Searle & Co.), Des Plaines, Ill.
The Ohmart Corporation, Cincinnati, Ohio
Tracerlab (Division of Laboratory for Electronics, Inc.), Waltham, Mass.
Robertshaw Controls Company, Anaheim, Calif.

Sales in this field are expected to continue growing at about the same rate as the rest of the nuclear instrument industry — about 15% per year — and could reach $40 to $50 million by 1976.

SELF-ILLUMINATING MATERIALS AND DEVICES

The use of self-illuminating materials on watches, signs, etc., continues to increase. The increased usage and interest are reflected by the fact that the number of curies of ^{3}H sold by the AEC has almost tripled in the past 3 years:

Fiscal year	Activity, curies	Sales
1963	60,000	$ 95,000
1964	112,000	125,000
1965	118,000	128,000
1966	171,000	196,000

The firms active in this field are:

American Atomics, Tucson, Ariz.
Canrad Precision Industries, Inc., New York
Tracerlab (Division of Laboratory for Electronics, Inc.), Waltham, Mass.
Luminous Products Corp., Boston, Mass.
Minnesota Mining & Manufacturing Co., St. Paul, Minn.
Radium Chemical Co., Inc., New York
United States Radium Corp., Morristown, N. J.

The AEC has received and acted on numerous petitions from a number of these firms to exempt from licensing the use of small amounts of ^{3}H, ^{147}Pm, and ^{85}Kr for various applications in the field of self-luminous materials and devices.

The AEC has exempted from licensing the use of ^{3}H or ^{147}Pm to provide self-illuminating automobile locks and ^{3}H for self-luminous dial timepieces. Additional items under consideration are thermostat

dials and pointers, radio dials and pointers, automobile gearshift quadrants, marine navigational instruments, glow lamps, safety markers for aircraft, highway signs, and airport runway markers.

In March 1965 the AEC published criterions that it will use in considering applications for use of radioactive materials in products intended for distribution to the general public.[13] In June 1965 the AEC simplified the format of its regulations[14] for the use of radioisotopes. The requirements applicable to all radioisotopes licensing were retained as Part 30 of Title 10 CFR, and new parts applicable to specific classes or categories of uses or users were issued as follows:

Part 31 — General Licenses for Certain Quantities of By-Product Material and By-Product Material Contained in Certain Items
Part 32 — Specific Licenses to Manufacture, Distribute, or Import Exempted and Generally Licensed Items Containing By-Product Material
Part 33 — Specific Licenses of Broad Scope for By-Product Material
Part 34 — Licenses for Radiography and Radiation Safety Requirements for Radiographic Operations
Part 35 — Human Uses of By-Product Material
Part 36 — Export and Import of By-Product Material

RADIOISOTOPIC POWER

Electric-power systems that operate on the energy released when radioisotopes decay are being developed for several specialized applications. Several units at power levels up to 60 watts(e) have been used successfully in remote terrestrial sites, and five units have been launched in the Department of Defense (DOD) Navigation Satellite Program.

Terrestrial. For terrestrial applications, power systems have been used in remote weather stations, navigation buoys, and as beacons on off-shore oil rigs. Additional systems are under development for:

1. Unattended sea surface and land surface systems (lighthouses, navigational buoys, automatic weather stations) for 1 to 5 years' life in the power range 10 to 200 watts
2. Extremely small, long-lived systems such as the cardiac pacemaker and the military Prescribed Action Link (PAL) systems in the power range of 1 μw to 1 watt
3. Deep-sea generators in the 10- to 1000-watt range

The AEC began developing an oceanographic nuclear power program in 1966 which includes both isotopic and reactor power sources. Isotopic power sources are expected to range from milliwatts to multikilowatts, and reactor power sources, to 2 Mw(e) or above, depending on the intended use. This program is integrated into the national oceanographic effort to ensure that the AEC's program is oriented to meet the demands of the national effort.

One of the most exciting and important new areas for the use of isotopic power devices is the development of a cardiac pacer. In 1965 the AEC selected NUMEC[15] to develop and test a highly compact unit with a power source capable of operating up to 10 years without being refueled. The power source will be fueled with ^{238}Pu. The potential market for these units has been estimated at about 10 thousand per year, with a ^{238}Pu fuel requirement of 3 to 4 kg per year.

Space. For space applications, four units, two of 2.7 watts and two of 25 watts, have been successfully launched in the TRANSIT 4 and subsequent DOD navigation satellites. The first, SNAP-3A, launched in 1961, now has been operating longer than any other orbiting space power system. Four of these units supplied the full design power during the whole life of the parent satellite system. A fifth unit at 25 watts was aboard a navigational satellite that failed to go into orbit. Evidence from fallout studies indicates the unit burned up safely in the upper atmosphere on reentry, as designed. Units are now being developed for the following:

1. Nimbus "B" satellite at 60 watts (two 30-watt modules on each satellite) to be launched in 1967
2. The 50-watt Apollo Lunar Surface Experiment Package (ALSEP) to be left on the lunar surface by the first Apollo astronauts
3. A family of DOD and NASA 90-day missions, at 500 watts, to be available for earliest possible flight in 1971, probably as part of a qualification test of a space satellite system

A thorough development program in advanced heat-source and power-conversion technology is also being carried out. A development program directed toward low-thrust high-specific-impulse isotope-powered thermal rockets for specialized space applications is also under way.

Industry. Isotopic power generation is a small but actively growing field. There are approximately 30 firms active in this area, and undoubtedly other firms will enter the field as markets develop.

The formation of a new company to develop isotopic power and heat sources, Sanders Nuclear Corporation, was announced in 1966 by Cambridge Nuclear Corporation and Sanders Associates, Inc.

During the year, NUMEC received an order for two ^{90}Sr-powered thermoelectric generators from the U. S. Naval Facilities Engineering Command.

The Martin Marietta Corp. announced fixed-price isotope-powered generators for sale. Their 25-watt ^{90}Sr-powered generators are intended for use in terrestrial applications. A deep-sea version is also available. The first Martin unit was sold to the Navy

Electronics Laboratory for installation on an island off the Alaskan coast for powering an oceanographic measuring station.

At present the federal government is the principal customer for these devices, although commercial markets, especially marine and terrestrial areas, and commercial space applications such as communications satellites are also promising. Radioisotopes such as ^{238}Pu, ^{90}Sr, and ^{210}Po are being used in these devices.

THE RADIATION PROCESSING INDUSTRY

In 1966, industry has evidenced a rapidly growing interest in commercial radiation processing applications. This field is divided into machine-produced radiation and isotope-produced radiation, each possessing some advantages and disadvantages over the other. Radiation processing applications using one or the other source include upgrading of plastics, curing of paints, upgrading of solid-state devices, sterilization of medical supplies, food preservation, production of wood–plastic combinations, production of shrinkable plastics, sterilization of parts for the space programs, production of biodegradable detergents, and synthesis of chemical compounds.

Table 3 lists the principal firms supplying accelerator equipment or services, while Table 4 lists suppliers of isotopic irradiators.

Rad-Mat, Inc., Gaithersburg, Md., was formed in 1966 to provide radiation processing services to the medical, plastics, food, electronics, and other industries. The company will use a 3-Mev Dynamitron accelerator, which will be the highest-powered source in industrial use.

The current commercial accelerator market, including Van de Graaff accelerators, Linacs, insulating core transformers, and Dynamitrons, is approximately $17 million annually. About three-fourths of this market is for Van de Graaff accelerators for research.

Table 3 FIRMS SUPPLYING ACCELERATORS OR SERVICES

Company	Accelerators	Services
Cyclotron Corp., Berkeley, Calif.	X	
General Electric Company, Milwaukee, Wis.	X	
Hughes Aircraft, Co., Culver City, Calif.	X	
Applied Radiation Corp., Walnut Creek, Calif.	X	X
Electra Process Industries, Inc., Burlington, Mass.		X
General Atomic (Division of General Dynamics), San Diego, Calif.	X	X
High Voltage Engineering Corp., Burlington, Mass.	X	
Electronized Chemicals Corp. (Subsidiary of High Voltage Engineering Corp.), Burlington, Mass.		X
Midwest Irradiation Center, Rockford, Ill.		X
Raycon, Windsor, Conn.		X
Radiation Dynamics, Inc., Westbury, N. Y.		X
Rad-Mat, Inc., Gaithersburg, Md.		X
Varian Associates, Palo Alto, Calif.	X	X
High Energy Process Company, New York		X
Radiation Polymer Corporation, La Grange, Ill.		X

Table 4 PRINCIPAL SUPPLIERS OF ISOTOPIC IRRADIATION FACILITIES

Company	Medical	Process	Research
Ameray Corp., Dover, N. J.			X
American Nuclear Corporation, Oak Ridge, Tenn.	X	X	X
The Budd Company, Phoenixville, Pa.	X		X
Gamma Process Company, Inc., New York		X	X
Iso Nuclear Corp., New York		X	
Martin Marietta Corp., Baltimore, Md.			X
Radiation Facilities, Inc., Lodi, N. J.	X	X	X
Radionics, Inc., Norristown, Pa.			X
Isotopes, Inc. (and subsidiaries), Westwood, N. J.	X	X	X
Lockheed-Georgia, Marietta, Ga.		X	X
The Babcock & Wilcox Co., Lynchburg, Va.		X	X
Nuclear Materials and Equipment Corp., Apollo, Pa.		X	X
Burns & Roe, Inc., New York		X	
Stone & Webster Engineering Corp., Boston, Mass.		X	
American Novawood Corporation, Lynchburg, Va.		X	
General Electric Company, San Jose, Calif.		X	
Westinghouse Electric Corporation, Pittsburgh, Pa.		X	

Large-scale (tonnage) private radioisotope process service irradiation facilities in the United States are practically nonexistent at present. While firms such as Lockheed-Georgia and American Novawood Corporation have substantial capacity for producing special products, no one firm offers a large-scale processing service. Irradiation, Inc., a subsidiary of Isotopes, Inc., was formed to introduce radiation processing to private industry and to aid businesses in bringing radiation processing into their production lines. The company has been awarded a contract to operate AEC's Portable Cesium Irradiator[16] for the Commission's program to demonstrate to food processors on a small scale the potential advantage of radiation processing.

An indication of the overall market for radiation processing and its growth may be obtained from the value of products processed: $20 million in 1963, $75 million in 1964, $150 million in 1965, and estimated over $250 million in 1966.

The irradiation processing of food is developing into one of the most important areas of applications of radiation. The AEC and U. S. Army programs for irradiation processing of food have been under way for many years, but only in the past 3 years have specific clearances for food been issued by the Food and Drug Administration.[17]

One of the major areas of interest, begun in 1961, is the development of irradiated wood–plastic materials. The AEC has sponsored a substantial amount of development work at West Virginia University, the

Air Reduction Company, and Lockheed-Georgia Company.[18] Arthur D. Little, Inc.,[19] Southern Interstate Nuclear Board,[20] Battelle–Northwest,[21] and Western New York Nuclear Research Center[22] have conducted market surveys to determine the steps necessary to achieve the earliest possible commercialization.

The Atomic Industrial Forum and the AEC sponsored an information meeting on irradiated wood–plastic materials in September 1965 for persons interested in the production, marketing, or use of these materials,[23] and in August 1966 the AEC released a report,[24] prepared by Vitro Engineering Co., on the design of a pilot-plant facility for the manufacture of these materials.

Under a demonstration program with Lockheed-Georgia, 80 companies cooperated in end-product testing programs to evaluate specific wood–plastic product applications.[25]

During the year, commercial prices for significant quantities of a variety of wood–plastic materials were announced by American Novawood Corporation, Lynchburg, Va., and Lockheed-Georgia Company, Marietta, Ga. These wood–plastic composites are being routinely manufactured in their radiation facilities. American Novawood is using ^{60}Co and Lockheed is using both their reactor and ^{60}Co. Louisiana-Radiant, Inc., Baton Rouge, La., has installed a ^{60}Co radiation source at the Louisiana State University's Nuclear Science Center for manufacturing wood–plastic samples, doing additional product development work, and training of company personnel. Two new companies, Gem Irradiated Wood, Inc., Dothan, Ala., and Iso Nuclear Corp., New York City, have announced plans to build plants for the commercial production of various wood–plastic composites. Both will be using ^{60}Co.

During 1966 the AEC solicited from industry expressions of interest in cooperative programs both for a wood–plastic materials demonstration facility and for a polyethylene radiation production facility. The industry responses generally indicated that the private capability now existing and planned might be adequate for development and production of these materials. In view of this, the AEC concluded that an AEC–industry cooperative program was not warranted at present but that AEC would review the situation after approximately a year to determine the extent of industrial progress. One private firm has plans to install a pilot plant for polyethylene production using ionizing radiation to initiate the reaction.

AEC Advisory Committee on Isotopes and Radiation Development

The present AEC Advisory Committee on Isotopes and Radiation Development, which was established in 1958, reviews AEC policies and the potential development of new applications of isotopes and radiation and makes appropriate recommendations for the Commission's consideration. This committee grew out of the Advisory Committee on Isotope Production and Distribution, which was formed in 1946 and terminated in 1958 when the Commission's regulatory and licensing activities were separated from the operating activities.

The composition of this committee has been gradually changed to bring in additional representatives from various segments of the isotope industry. At the committee's suggestion, the membership was reduced from 23 to 18 during 1966. The present membership is:

Chairman, John L. Kuranz, Nuclear-Chicago Corporation
Vice Chairman, John W. Landis, The Babcock & Wilcox Co.
AEC Member, Dr. George M. Kavanagh, Assistant General Manager for Reactors

Subcommittee on Commercial Activities
Chairman, Howard K. Nason, Monsanto Research Corporation
Dr. John C. Brantley, Nuclear Science and Engineering Corporation
David E. Trumbull, General Electric Company
Dr. Charles Artandi, Ethicon, Inc.

Subcommittee on Process Radiation
Chairman, Dr. Willard P. Conner, Hercules Research Center
Dr. Vivian T. Stannett, North Carolina State University, Raleigh
Dr. David E. Harmer, Dow Chemical Co.
E. Robert Kinney, Gortons of Gloucester, Inc.

Subcommittee on Radioisotope Power
Chairman, Dr. Leonard Reiffel, National Aeronautics and Space Administration
Dr. Nathan W. Snyder, Georgia Institute of Technology
Dr. Thomas E. Hicks, University of California at Los Angeles
Dr. Ernst Stuhlinger, Department of the Army, Marshall Space Flight Center

Subcommittee on Radiation Systems
Chairman, Dr. Rodman A. Sharp, Beckman Instruments, Inc.
Henry R. Chope, Industrial Nucleonics Corp.
Dr. Paul M. Stier, Union Carbide Corporation
Dr. Donald W. Pritchard, Johns Hopkins University

In 1965 a study was conducted for the AEC by Dr. Lauchlin M. Currie, then Chairman of the AEC Advisory Committee on Isotopes and Radiation Development, and O. M. Bizzell, of the Division of Isotopes Development, to determine the extent of industrial development of isotopes and radiation, to identify factors that limited their use, and to obtain recommendations on how AEC might better help this portion of the atomic energy program.[26] Recommendations were submitted to the Commission, and all have been acted upon or are in the process of being handled.

(RHL)

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Solvent Extraction of Fission Products

By F. J. Miller

Part I. Strontium

Abstract: The extraction of useful radioisotopes from solutions accumulated in the processing of spent fuel elements during recovery of uranium and plutonium is important in the economic reclamation of fission products and storage of the process solutions. The basic phenomena of solvent extraction are reviewed here, and the necessary equipment is discussed briefly. As a typical example of the reclamation of a commercially useful radioisotope, the solvent extraction of ^{90}Sr with di-(2-ethylhexyl) phosphoric acid diluted with kerosene is described in detail.

Solvent extraction is an essential process both in the economical reclamation of potentially valuable isotopes from spent reactor fuel and in the reduction of costs of waste storage. Rupp[1] has pointed out that with the proliferation of nuclear-fueled power-generating facilities—7 ordered[2] in 1966, bringing

the total installed or on order to 26 — the concomitant production of fission products from processing of spent reactor fuel elements will increase enormously.

Safe storage of the long-lived radioactive by-products of nuclear reactors for hundreds of years is required to prevent contamination of water and food supplies. Providing the required amount of storage space is costly, and much attention is presently being given to reducing costs by treatment of the by-product material or by alternative means of storage. Extraction of the radioactive isotopes from the bulk waste solution and conversion of the extract to solids would permit secure storage in a greatly reduced space. In particular, the solvent extraction and encapsulation of ^{90}Sr and ^{137}Cs would lessen the difficulties associated with the handling and storage of fission-product materials.[3]

The operating costs of nuclear-fueled power plants can also be reduced if the same energetic nuclides that pose the greatest problem in the storage of fission products can be put to useful purposes such as continuous sources of heat, electricity, and specific types of radiation. The isotopes of prime interest are ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm (Ref. 4). Although isotopic power sources have certain unique qualities, they must compete in cost with other sources of energy. Since a major portion of the costs of isotopic power sources is that of reclaiming the isotopes from the bulk process solutions, these production costs must be minimized.[5]

Solvent extraction has long demonstrated its applicability to the separation and purification of radioactive isotopes. Particular advantages are economy, adaptability to the remote-handling operations required in the handling of intensely radioactive material, and applicability to either continuous or batch processing for the separation, purification, and concentration of a desired component that may be present in very low concentration.

This discussion will cover the basic elements of liquid–liquid solvent extraction and the extraction of ^{90}Sr as a typical example of the extraction of a radioisotope. Future articles will cover in specific detail the solvent extraction of other isotopes that are contained in process solutions generated in the processing of spent nuclear fuels to recover uranium and plutonium.

Liquid-Liquid Solvent Extraction

Liquid–liquid solvent extraction is a process in which two mutually insoluble liquids are brought together for the purpose of transferring a dissolved substance in one liquid to the other liquid. The two immiscible liquids may be two liquid metals, a liquid metal and a molten salt, an aqueous solution and an organic solvent, or any of a large variety of other systems. In this discussion the two liquids considered are an aqueous phase and an organic phase that are essentially mutually insoluble; the dissolved substance is an inorganic (usually metal) ion. The ion distributes itself between the two phases in accordance with the Nernst partition isotherm[6] theory, which states that the ratio of the concentrations of the dissolved substance in the two phases at a particular temperature is a constant if the solute has the same molecular weight in the two phases.

If the metal ion can form an uncharged species either with the organic solvent or with an agent added to the aqueous–organic system, then the uncharged species will distribute itself preferentially into the organic phase. The degree of preference[7] is expressed as a distribution coefficient, D:

$$D = \frac{\text{concentration of the species in the organic phase}}{\text{concentration of the species in the aqueous phase}}$$

Implicit in this statement is the ability of the solvent extraction process to function over a wide range of concentrations. Thus, if 95% of the dissolved material in an aqueous solution is transferred in one extraction into an organic phase, then a second extraction with fresh organic solvent will extract 95% of the 5% remaining in the aqueous phase. The process may be repeated ad infinitum until the amount remaining in the aqueous phase is below the limits of detection, provided that the slight mutual solubility of the phases and the physical carry-over of the aqueous phase have only insignificant effect.

METHODS OF INCREASING THE SOLUBILITY OF THE ION IN THE ORGANIC PHASE

The extractable component of an aqueous solution may be made more soluble in the organic phase than in the aqueous phase by means that vary in degree of sophistication. The simplest method is to change the oxidation state of the ion so that an uncharged molecule results. Thus, if I^- is oxidized to I_2 by the addition of an oxidant to the aqueous solution, the I_2 produced may be readily extracted into a nonpolar solvent such as carbon tetrachloride.

A salting-out agent may be used to reduce the solubility of the ion in the aqueous phase. Salting-out agents are ionic salts that dissolve in the aqueous phase in high concentration. The heavy concentration of the salting agent can shift the chemical equilibrium so that the ion of the desired metal is forced into an un-ionized combination. Uranyl nitrate, for example, in dilute concentration is not extracted into an organic phase from an acid solution but is made readily extractable by the addition of a salting-out agent, e.g., $LiNO_3$, $Al(NO_3)_3$, or $Ca(NO_3)_2$.

The hydrogen-ion concentration of the aqueous solution may be varied so that, in a manner similar to the use of a salting-out agent, the chemical equilibrium is shifted and an un-ionized species of the metal component is produced. Nitric acid, which itself is readily extractable into many organic sol-

vents, can act both as a solvating acid and as a salting-out agent. In the extraction of uranyl nitrate into *n*-butyl ether, for example, the nitric acid actually competes with the metal salt in the process of solution into the organic phase. The distribution coefficient of uranyl nitrate thus varies with the acid concentration. An acid concentration can be selected that permits effective extraction but represses hydrolytic precipitation of the uranyl ion. Ions of the same metal but in different oxidation states differ also in hydrolytic behavior; the difference in degree of hydrolysis is reflected in the extraction process. Entire solvent extraction schemes such as the "Redox" process[8] are based on the interplay between the hydrogen-ion concentration and the degree of hydration of the various oxidation states of uranium, plutonium, and fission products.

Frequently an organic compound that will react with the metal ion to form a nonpolar organic-soluble metallo-organic complex is added to the aqueous–organic system. The solubility of the nonpolar complex is much greater in the organic than in the aqueous phase. The organic additive may be a chelating compound, a liquid amine, an organic phosphate, or a weak organic acid that can function as a liquid ion exchanger. Examples of such additives are given by Morrison and Freiser[9] and in other selected references.[6,10,11]

In many extraction processes, successful separations are achieved by use of additives that form water-soluble rather than organic-soluble complexes with metal ions. Typical complexants are the organic acids—tartaric and citric—and chelating compounds such as the sodium salt of ethylenediaminetetraacetic acid (EDTA). Tartaric and citric acids prevent hydrolysis and precipitation, at elevated *p*H, of easily hydrolyzable ions and hinder their extraction by forming with these ions a stronger, more water-soluble complex than is formed with the organic-soluble extractant.

Buffering action, which is required in many processes to maintain a fixed *p*H, is also provided by a mixture of the weak organic acid with its sodium salt. The chelating agents form negatively charged, water-soluble species with metal ions but require the use of an additional buffering agent.

Table 1 SYNERGISTIC EFFECT OF MIXED EXTRACTANTS

Aqueous Phase: 0.5*M* SO_4^{2-}, *p*H 1, and 0.004*M* U(VI)
Organic Phase: Extractant in Kerosene
Volume Ratio of Phases: 1:1 at 25°C

Extractant	Concentration, *M*	Extractant alone, D.C.*	Combined with 0.1*M* D2EHP, D.C.*
Di-(2-ethylhexyl) phosphoric acid (D2EHP)	0.1	135	-
Phosphate			
Tri-*n*-butyl (TBP)	0.1	0.0002	470
Tri-2-ethylhexyl	0.1	0.0002	270
Phosphonates			
Di-*n*-butyl-*n*-butyl	0.1	0.0002	1700
Di-*n*-amyl-*n*-amyl	0.1	0.0003	2000
Di-*n*-hexyl-*n*-hexyl	0.1	0.0004	2200
Di-2-ethylhexyl-2-ethylhexyl	0.1	0.0002	870
Phosphinates			
n-Butyl-di-*n*-butyl	0.1	0.002	3500
n-Butyl-di-*n*-hexyl	0.1	0.002	3500
Phosphine oxides			
Tri-*n*-butyl	0.05	0.0025	7000
Tri-*n*-octyl	0.1	0.06	3500
Tri-2-ethylhexyl	0.1	0.02	650

*D.C., distribution coefficient.

Occasionally two organic additives, when used separately, extract the metal ion poorly; however, the combined action of the two can result in a very efficient extraction. The joint action of two agents to enhance the effect of either used singly is referred to as "synergism." Adapted from the medical vocabulary, the term "synergism" was first used by Coleman et al.[12] Vdovenko[11] gives an excellent discussion of synergism and tabulates several synergistic pairs (Table 1). Baes[13] has also discussed synergism, especially in regard to organophosphate systems.

Ions of different metals complexed with the same organic compound may not transfer from the aqueous to the organic phase at the same rate. Advantage may be taken of the kinetic factor to separate ions of different metals by varying the contact time of the two phases. In solvent extraction processes the flow rates become important parameters. Careful adjustment of the flow rates provides a contact time sufficient to extract a desired component with a minimum carry-over of an undesired ion. Wischow and Horner[14] recognized and used the different transfer rates of Sr–D2EHP [di-(2-ethylhexyl) phosphoric acid] and Fe–D2EHP from the aqueous to the organic phase to separate strontium from iron contamination.

Selection of Diluent

In process chemistry the complexant is usually diluted with kerosene or a similar proprietary solvent in preference to more expensive nonpolar organic solvents that are used in analytical-chemistry procedures. Economy is not the sole reason for the choice of diluent.[15] The diluent itself is directly involved in the extraction process,[16] and the degree and mode of solvation of the extractable species in it need to be considered, especially since the solvating ability of the diluent determines to a considerable extent the required volume ratio of the phases. The viscosity and density of the diluent also are factors in determining its choice. The diluent should be chemically stable under process conditions and in an intense radiation field. Oxidation by a relatively high nitric acid content of the aqueous phase can cause rapid deterioration of an otherwise suitable diluent. If the diluent can be decomposed by radiation, the *p*H of the solution could shift with consequent impairment of the extraction. Polymerized components of the diluent may cause further difficulty by creating emulsions or by plugging equipment lines. The choice of kerosene is based primarily on cost, chemical and radiation stability, and ability to solvate the extractable species.

Precautions must be taken to guard against third-phase formation in kerosene-type solvents. Third-phase formation in an aqueous–organic system results when two distinct phases are formed in the organic layer in addition to the aqueous phase. The phenomenon occurs at a fixed *p*H and anion concentration and is directly influenced by the limited solubility of the organic extractant in the diluent. One phase of the organic diluent becomes loaded with the metallo-organic extract; the other remains relatively pure solvent. As a consequence of third-phase formation, extraction efficiency is decreased, and the process may cease altogether.

The D2EHP–kerosene–aqueous system, as an instance, is prone to third-phase formation because of the limited solubility of D2EHP in kerosene. When D2EHP, diluted with kerosene, is used as an extractant, third-phase formation can be repressed by adding tributyl phosphate (TBP) to the system, which serves to increase the solubility of D2EHP in the diluent. The combined use of TBP and D2EHP increases the efficiency of the extraction but is not a true case of synergistic action. The putative synergistic action of TBP–D2EHP is actually the result of maintenance of D2EHP solubility in the organic phase. The combination of TBP and D2EHP is used in the extraction of Sr^{2+}.

SCRUBBING AND STRIPPING

Scrubbing and stripping steps are integral to a complete cycle for solvent extraction and recovery of a fission product. In a scrubbing step the organic solvent that contains the extractant combined with the metal ion is placed in contact with an aqueous phase suitably adjusted in *p*H and possibly containing an additional water-soluble complexing agent. The scrub solution is used to selectively remove an undesired component from the organic solution, for instance, to decrease the sodium content of a Sr–D2EHP solution in the kerosene diluent.

Stripping requires placing the organic phase in contact with an aqueous phase so adjusted in *p*H and other possible additives that the metal–organic species is dissociated and the desired metal ion transferred to the aqueous phase. The economy of the extraction process is based largely on the efficiency of the stripping step. The organic solution containing the extractant is not destroyed or consumed in use but can be recycled continuously; only slight amounts of fresh material need be added. The cost of the extractant and organic diluent is spread over a large number of repetitive extraction cycles and becomes economically justifiable. Further, if the stripping process is efficient, only a small volume of aqueous stripping solution is required. The final product is obtained as a nearly pure material in a highly concentrated solution.

Solvent extraction is a topic of importance through all of chemistry. Benedict and Pigford[7] discuss solvent extraction as an engineering process; Morrison and Freiser,[9] as an analytical tool; and Irving,[6] as a means for the elucidation of inorganic chemistry problems. These and other workers[17–20] in the field of solvent extraction chemistry should be consulted for details.

EQUIPMENT

Basically the only equipment required for liquid–liquid solvent extraction is a stoppered container in which the two phases may be shaken together until the maximum extraction possible has been achieved (i.e., until equilibrium has been reached). To separate components that have only slightly different distribution coefficients, multiple extractions are necessary, and this is usually done as a continuous process. The simple separatory funnel used in the laboratory for a single-pass extraction is too cumbersome and ill-adapted for continuous extraction. Accordingly, many different types of continuous extractors have been devised for laboratory use.

The most effective of the early devices was that of Craig,[6] who developed one of the first countercurrent flow machines in which the sample stream and the organic moved from stage to stage in opposite directions. The equipment consisted of a number of glass vessels so arranged that, after an equilibration period, the solvent was automatically transferred from one vessel to another. Using a battery of vessels in a single frame, Craig was able to separate components of very complex organic mixtures.

Following Craig's work, various types of equipment have been developed which are more suitable for large-scale process work. The mixer–settler is a compact box-shaped unit divided into two chambers—a mixing chamber and a settling chamber. The organic solvent and the aqueous sample stream enter the mixing chamber, which contains a rotating impeller and baffle plates. After being thoroughly mixed, the combined streams enter the settling chamber. The mixture becomes quiescent and divides into two phases, which exit through top and bottom ports. A number of such units may be connected in series to provide the required number of extraction stages. A typical mixer–settler setup as installed in a hot cell is seen in Fig. 1.

Pulse-flow columns are frequently used in process extraction. They are vertical pipes that contain a large number of horizontally mounted perforated stainless-steel plates. Mixing takes place at the interface created by each horizontal plate. The organic and aqueous streams are fed into the column in opposite directions by pulse-feed pumps. The alternate surge and retraction of the pulsed streams creates turbulence in the column, thus inducing better mixing at the plates.

The packed column is another of the more widely used types of extraction equipment. It is a vertical pipe packed with a coarsely divided material such as small porcelain or stainless-steel rings. The packing material is inert and serves only to provide a very large surface area which is thinly wetted by the flowing streams. The sample stream and the solvent stream enter in opposite directions and flow counter to each other. The pipes may be connected to each other in cascade to provide an adequate number of stages.

A more recent development is the Luwesta centrifugal extractor.[21] In this unit the feed and solvent liquids are intensively mixed by simultaneous flow through alternate parallel nozzles to establish extraction equilibrium, which is followed by a phase separation in the centrifugal separator bowl. This extractor, because of its relatively small holdup volume and residence time, is particularly suitable

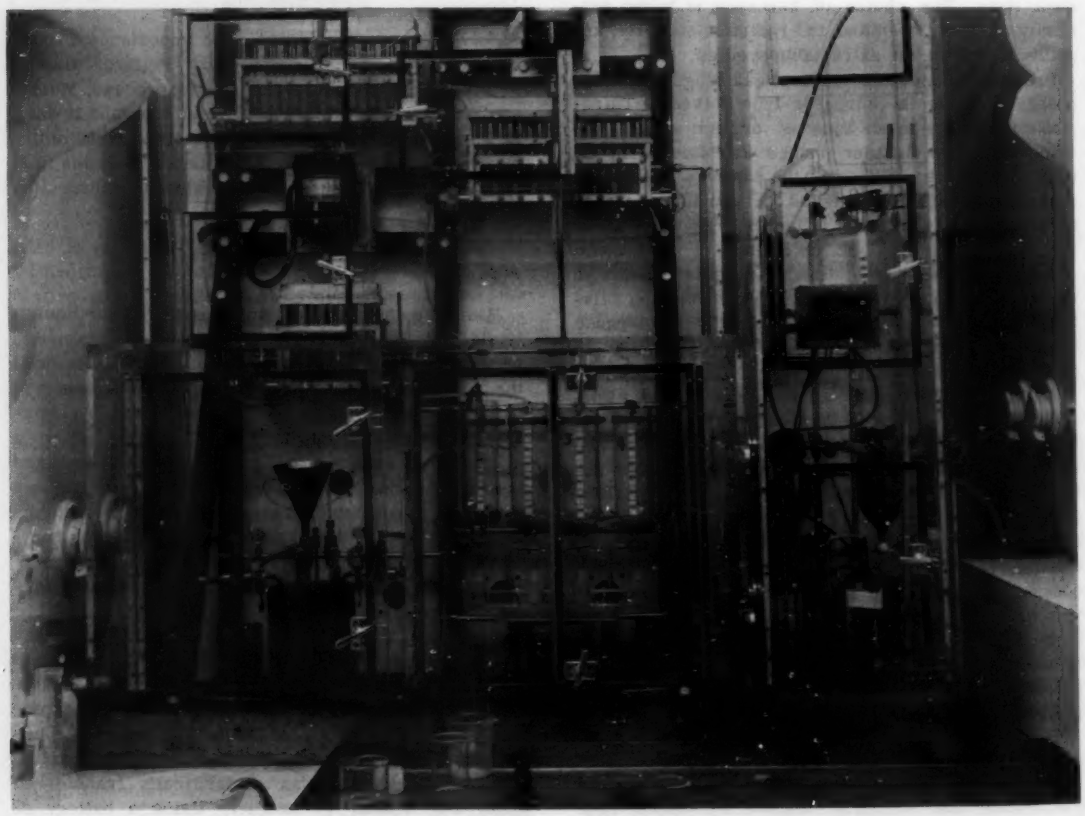

Fig. 1 Typical remote-handling extraction setup.

for chemically and thermally unstable substances, although it has not yet come into use in the nuclear industry. Other new developments in extractor equipment available in the United States were covered by Akell,[22] and those abroad, by Reman,[21] at a solvent extraction symposium.

Extraction of ^{90}Sr from Fission-Product Process Solutions

Specific and detailed methods for the recovery and purification of fission products will be covered in an Oak Ridge National Laboratory (ORNL) report by Crouse and Horner,[23] but the extraction of ^{90}Sr is typical of the separation of a commercially important isotope from fission-product process solutions. The ^{90}Sr is contained in process solutions from the Purex[24] process, in which uranium and plutonium are recovered from a nitric acid solution of spent fuel elements by extraction with TBP in kerosene. Nitric acid acts both as the solvent acid and as a salting-out agent, thereby eliminating the $Al(NO_3)_3$ used in the earlier Redox process. The aqueous waste, after extraction, is concentrated by evaporation and then pumped to storage tanks.

Purex wastes are not all identical because various methods are used to decrease the nitric acid content of the stored solutions. Formaldehyde, sugars, or other additives are used to reduce nitric acid to gaseous nitrous oxides, which may be sparged from the solution. The classification of Purex wastes is further complicated by the consideration of only the supernatant liquid from the storage tanks in some cases, and of the sludge from the tank bottoms in others. In all cases, however, a typical Purex-waste solution can be assumed to contain, in addition to nitric acid, the cations H^+, Na^+, Fe^{3+}, Al^{3+}, Cr^{3+}, Ni^{2+}, UO_2^{2+}, Ce^{4+}, Si^{4+}, Cs^+, $Ru^{(?)}$, Zr^{4+}, and Nb^{4+}; rare-earth ions; and the anions NO_3^-, SO_4^{2-}, PO_4^{3-}, and $C_4H_4O_6^{2-}$. One or more prior processing steps may be required in conjunction with solvent extraction, or solvent extraction may be used only as a final purification step. Wischow and Horner[14] in 1962 discussed the recovery of strontium and rare earths from Purex wastes by solvent extraction alone. Horner et al.[25] at Oak Ridge National Laboratory, Peppard and Mason[16] at Argonne National Laboratory, and Cooley and Richardson[26] at Hanford Laboratories have presented more recent studies of the solvent-extraction process.

Although other extractants have been used[27] for ^{90}Sr, D2EHP is the one most commonly used. The acid is classed as weak, the hydrogen ion being strongly associated with the acid radical in acid solution. The acid has been assumed to function as a liquid ion exchanger by replacement of the hydrogen ion with a metallic cation at an appropriate *p*H. The *p*H at which a cation will replace the hydrogen ion depends upon the properties of the specific cation. If NaOH is used to raise the *p*H, then the replacement process must be considered as a competiton between the H^+, Na^+, and the other cations in solution. Baes, Zingaro, and Coleman[28] describe the reaction as

$$M^{n+}_{(aq)} + nHA_{(o)} \rightleftharpoons MA_{n\,(o)} + nH^+_{(aq)} \qquad (1)$$

where M = a metal ion
HA = the reacting acid
H^+ = the hydrogen ion
n = the number of moles of reactant
(o) = the organic phase
(aq) = the aqueous phase

The actual equilibrium is much more complicated since the alkyl phosphoric acid in the uncombined state exists usually as a dimer or higher state of polymerization.

In the metal complex MA_n of Eq. 1, the anion A is not a simple radical but a complex polymer. To represent the reaction of the cation as a simple ion-exchange reaction is to oversimplify almost to the point of being misleading. The structure of the complex is much more likely to be that of a chelate. Nonetheless, as the *p*H of the solution is raised by the addition of NaOH, equilibria are set up in which the H^+ is replaced by Na^+, which in turn is replaceable by Sr^{2+}. Wischow and Horner[14] represented the mechanism by the reactions

$$2\,Na^+_{(aq)} + [HA]_{2\,(o)} \rightleftharpoons 2NaA_{(o)} + 2H^+_{(aq)} \qquad (2)$$

$$Sr^{2+}_{(aq)} + 2[HA]_{2\,(o)} \rightleftharpoons SrA_2(HA)_{2\,(o)} + 2H^+_{(aq)} \qquad (3)$$

$$Sr^{2+}_{(aq)} + 2NaA_{(o)} \rightleftharpoons SrA_{2\,(o)} + 2Na^+_{(aq)} \qquad (4)$$

If the above set of equations is accepted as a working model of the system, then any change in the concentration of H^+, Na^+, and Sr^{2+} will affect the complexation and extraction of strontium. Consequently the system must be studied to determine optimum conditions for extraction of the desired ion and, conversely, to minimize coextraction of other ions. McDowell and Coleman[29] in 1965 reported a detailed study of the mechanisms involved in the extraction of Na^+ and Sr^{2+} with D2EHP.

Tartaric or citric acid may be added to the aqueous–organic system to act simultaneously as a water-soluble complexing agent and *p*H buffer. These acids form complexes with metallic cations, as does D2EHP, but the complex is negatively charged and thus water soluble. Sodium EDTA and certain other chelating agents also form water-soluble complexes with metallic cations but do not buffer the solution because of the insolubility of EDTA in acid solutions. Consequently a buffer such as sodium acetate–acetic acid is added to stabilize the *p*H. Wischow and Horner[14] showed that strontium and rare earths could be extracted from a neutral sodium nitrate solution with D2EHP but could not be extracted from

acidic or neutralized Purex-process solutions because of iron interference. By adding tartrate ion to the solution, the *p*H was buffered to a constant value and the iron effectively tied up in a water-soluble complex and retained in the aqueous phase. Strontium was also complexed by the tartrate ion but not so strongly as iron. The degree of complexing depended on the *p*H of the solution. An optimum *p*H was found at which the iron was held in the aqueous phase by the tartrate complex and the strontium transferred to the organic phase by complexing with D2EHP. Essentially, this process may be regarded as a rivalry between the D2EHP and the tartrate ion for the strontium ion, and conditions had to be established which favored complexation with D2EHP.

Although the bulk of the iron was held in the aqueous phase by the tartrate ion, some was transferred to the organic phase. The amount was decreased by taking advantage of the kinetics of the transfer-rate process. The rate of transfer into the organic phase of the Fe–D2EHP complex was known to be much slower than that of the Sr–D2EHP. By minimizing the time during which the aqueous and organic phases were in contact with each other, the Sr–D2EHP transfer was favored over that of iron.

The rare earths are extracted from Purex-process streams equally as well as strontium in the *p*H range 5 to 6. In the *p*H range 2 to 3, however, strontium extraction is negligible, but the rare earths are extracted readily. Thus rare earths can be separated and recovered by consecutive extractions with an intervening *p*H adjustment. Alternatively, rare earths and strontium can be extracted simultaneously at *p*H 5 to 6 and then separated from each other by a second-cycle extraction.

TYPICAL PROCESS

A typical solvent extraction process has been described by Schulz, Mendel, and Richardson[30] and by Cooley and Richardson[26] from Hanford plant Purex-process solutions. The work is based on earlier developmental studies carried out at ORNL.[23] The process was developed to provide megacurie quantities of highly purified radiostrontium for use in a thermoelectric-power generator. The composition of the typical plant feed solution is shown in the left-hand column of Table 2; laboratory development was carried out with a synthetic solution made up as shown in the right-hand column of Table 2.

Briefly, as shown in the flow diagram of Fig. 2, active strontium and inert calcium were extracted by D2EHP diluted with a kerosene-type hydrocarbon fortified with TBP from an aqueous feed solution buffered to *p*H 4.7 with sodium acetate–acetic acid. Either NaEDTA or pentasodium diethylenetriaminepentaacetate (NaDTPA) was added to the feed solution. By forming water-soluble complexes with iron, lead, and the fission products ^{95}Zr–^{95}Nb and ^{106}Ru–^{106}Rh, the complexing agents almost completely suppressed the extraction of these elements. Some separation from fission-product rare earths was also obtained in this step. Most of the sodium that was coextracted with strontium into the organic phase was removed by an aqueous scrub solution that was 0.6*M* in citric acid at a *p*H of 2.6. In a succeeding column, strontium was stripped from the organic phase by contact with an aqueous stream 1*M* in citric acid at a *p*H of 1.5 to 3.5. The citric acid content and the *p*H were critical factors that had to be closely controlled so that strontium but not calcium was stripped from the organic phase. Further purification from rare earths, especially Ce^{4+}, was obtained by additional solvent extraction cycles or by an ion-exchange cycle. Processing costs were 0.6 to 0.7 cent/curie of ^{90}Sr. Crouse and Horner[23] discuss a very similar solvent extraction for the ^{90}Sr procedure. However, it should not be inferred that solvent extraction is the universal answer to all ^{90}Sr processing problems. These authors also present processes in which precipitation, ion exchange, and solvent extraction are combined to achieve an optimum process dependent upon the nature of the starting feed.

Table 2 COMPOSITION OF CRUDE ^{90}Sr CONCENTRATE SOLUTION

Typical plant concentrate			Laboratory concentrate	
Component	Concentration, *M*	Activity, curies/liter	Component	Concentration, *M*
HNO_3	0.80		HNO_3	1.0
$Fe(NO_3)_3$	0.025		$Fe(NO_3)_3$	0.10
$Pb(NO_3)_2$	0.020		$Pb(NO_3)_2$	0.004
$Ca(NO_3)_2$	0.005		$Ca(NO_3)_2$	0.003
^{90}Sr		17 – 24	$La(NO_3)_3$	0.0025
^{144}Ce		40 – 260	$Sr(NO_3)_2$	0.0025
^{95}Zr–^{95}Nb		0.5 – 40	$Ce(NO_3)_3$	0.0001
^{106}Ru–^{106}Rh		0.1 – 2.0	$ZrO(NO_3)_2$	0.001
			$RuCl_3$	0.00002

McHenry and Posey's work[31] has been used as a basis for the separation of strontium from calcium in process operations. In one such process,[23] solvent extraction was used as the final clean-up step for the separation of strontium from calcium. In the feed to the extraction cycle, the Ca/Sr ratio ran as high as 20/1. By use of a multiple-stage, countercurrent extraction with D2EHP, 2-ethylhexanol, and acetate buffer, strontium was recovered in amounts greater than 99% with an amount of calcium present that was below the limits of analytical detection.

A flow sheet as shown in Fig. 2 is deceptively simple in appearance. The concentrations of reagents and the flow rates as plainly stated in the flow sheet tend to disguise the intricacy of the chemistry involved in the process. An effective solvent extraction

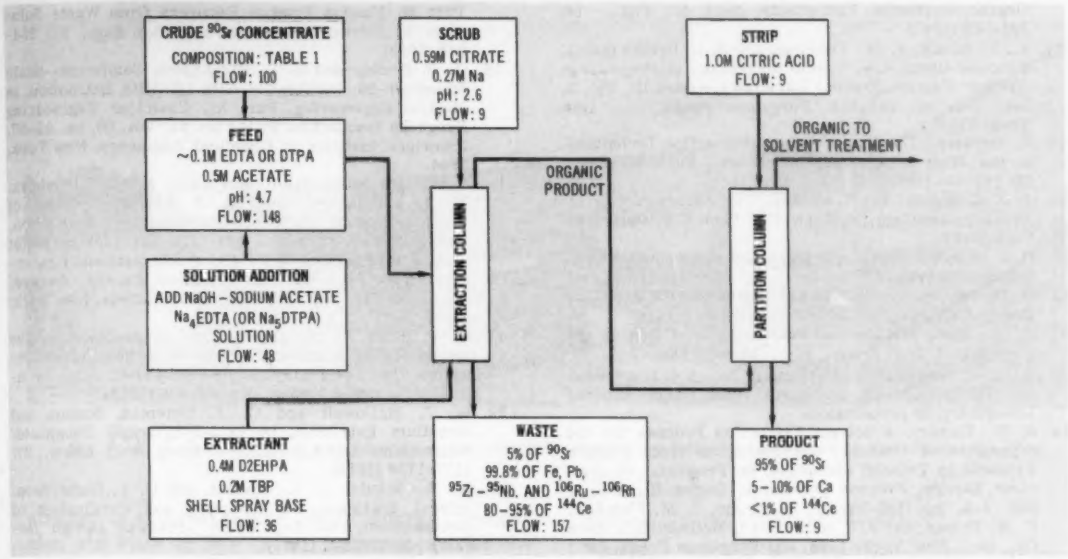

Fig. 2 Chemical flow sheet used in the recovery and purification of about 1 Mc of ^{90}Sr.

scheme requires detailed knowledge of complex constants for both organic and water-soluble complex compounds, the effect of *p*H on formation of the complexes, the kinetics of the transfer rate between the phases, radiation effect on organic solvents and reagents, and other possible variables. All the variables are interrelated and mutually dependent. The test of the process chemist's and engineer's skill is in balancing exactly all variables to produce a seemingly simple process into which the raw feed enters and from which the highly purified product emerges. The widespread use of solvent extraction in processing of radioactive material attests to the skill of the engineer in application of the fundamental knowledge supplied by the laboratory chemist.

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Radiopharmaceutical Production at Squibb

By Angelo Virgona*

Abstract: The Squibb radiopharmaceutical processing facility in New Brunswick, N. J., is a 27,500-sq ft building specially equipped for processing radioactive drug materials.

Squibb entered the radiopharmaceutical field in early 1957—at a time that the advent of a number of large nuclear reactors had ensured an abundant supply of radioisotopes. Its first three products were sodium iodide-^{131}I (Iodotope†), sodium phosphate-^{32}P (Phosphotope†), and ^{198}Au (Aureotope†). Fourteen additional products were added in 1958, and, each year since, the product line has been expanded until in 1967 more than sixty items are available.

At the time of Squibb's entry into the field, practically the only source of large quantities of radioisotopes was Oak Ridge National Laboratory, which is operated for the federal government by Union Carbide Corporation. However, a stimulus for commercial production of radioisotopes was provided by the decision of the federal government to discontinue production of an isotope when it became available from private industry at reasonable cost and comparable purity. At present the bulk of the isotopes used for radiopharmaceutical manufacture is supplied by private concerns, e.g., Union Carbide Corporation at Tuxedo, N. Y., General Electric Company (Vallecitos) at Pleasanton, Calif., and Nuclear Science and Engineering Corporation at Pittsburgh, Pa., with Oak Ridge supplying only those isotopes not yet commercially available. A large nondomestic supplier is Atomic Energy of Canada Limited.

*Present address: Union Carbide Corporation, Sterling Forest Research Center, Tuxedo, N. Y.

†Squibb trademark.

Squibb Production Facilities

The initial manufacturing area, 3500 sq ft in 1957, was expanded to 4000 sq ft in 1959. However, by early 1962, facilities were no longer adequate to meet the demands of the rapidly expanding market. Therefore, to provide for expected future growth as well as for immediate requirements, $2 million was approved for the construction of a new one-story building near New Brunswick, N. J.

The new building (Fig. 1), with an area of about 27,500 sq ft, was completed in January 1964. Construction is of reinforced concrete and steel, faced with tile, with special concrete where required for radiation shielding. All processing areas were con-

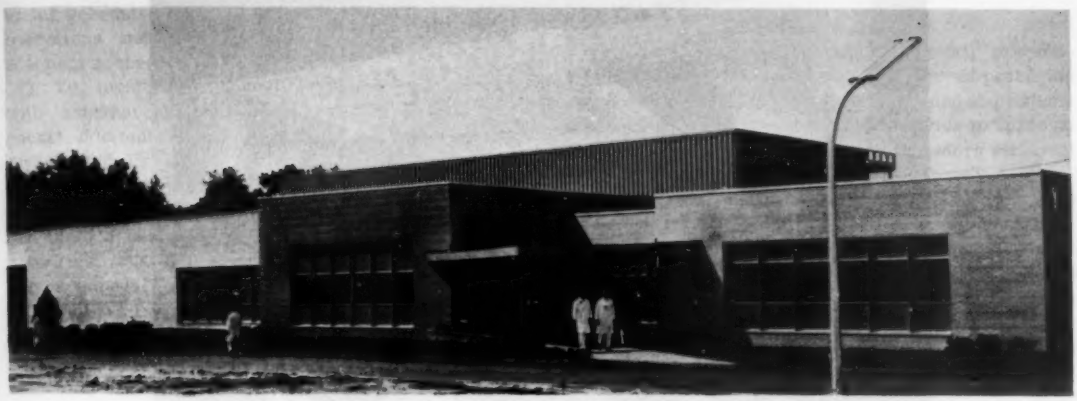


Fig. 1 E. R. Squibb & Sons' new $2 million radiopharmaceutical manufacturing laboratory in New Brunswick, N. J.

structed in accordance with Atomic Energy Commission and state of New Jersey requirements for the handling of radioisotopes to protect personnel from overexposure to radioactive materials. The $2 million total cost of the new building may be broken down as follows:

Building proper	29.5%
Special heavy-duty laboratory tables, hoods, and equipment	20.0%
Special air conditioning, ventilating, and scrubbers for exhaust air	16.0%
Electrical system	12.0%
Piping, services, and plumbing	10.7%
Special high-activity-level caves	9.3%
Holding tanks, valves, and controls	2.0%
Miscellaneous (landscaping and roads)	0.5%

BUILDING

The walls are painted with a urethane-base paint, and the floors are covered with vinyl linoleum to minimize absorption of radioactive substances and simplify decontamination. Cool-shade glass was used for all outside windows. The special barytes concrete used for shielding the counting rooms and the high-activity-level caves has a density twice that of ordinary concrete and about 40% that of lead. Three thousand cubic feet—750 thousand pounds—of this concrete was used.

The building is equipped with an intercom and pneumatic message system for more efficient supervisory control and rapid processing of orders. Teletype machines in the order-processing area are connected to 15 Squibb branches.

Because of the heavy lead linings required for radioactive shielding, hoods (Fig. 2) were specially reinforced. Stainless steel was used extensively for ease of cleaning and minimum maintenance.

The building is completely protected against lighting failure with a battery-operated emergency lighting system.

AIR CONDITIONING AND VENTILATION

Except for the machine rooms, the building is fully air-conditioned by a one-pass rather than a recirculating system because of the radioactive materials handled. Air is supplied through Cambridge filters, which remove microscopic particles and bacteria, to the rooms where sterile products are prepared. The air intake is protected by prefilters, carbon filters, and precipitrons to prevent organic fumes from other industrial sites from passing into the building and poisoning the carbon filters in the exhaust system, which are designed primarily to prevent escape of any radioactive contamination. The exhaust system also has two electrostatic precipitrons, one before and one after the filters, the total filter area being approximately 300 sq ft. Thirty thousand cubic feet per minute is handled through the system. Humidity and temperature are also carefully controlled.

SPECIAL HIGH-ACTIVITY-LEVEL CAVES

Three fully equipped rooms contain high-activity-level caves, where operations are performed with the latest type manipulators (Fig. 3). A fourth room provides for future expansion. The caves, which are made from high-density concrete, lead, and leaded-glass windows, were designed to handle up to 500 curies of activity of 0.5 Mev energy. Each cave is divided into two compartments, separated by a biparting door that is operated remotely. In the larger compartment are performed the actual manipulations of subdividing and diluting of the highly radioactive materials used in compounding a drug. The bulk-radioisotope containers are introduced into the smaller compartment by a forklift truck, the door is closed, and the containers are opened by remote control. Then the biparting doors are opened and a container, by remote means, is moved into the larger

Fig. 2 Partial view of one of the Medotope prescription filling areas. The hood at right is specially reinforced to carry the heavy shielding load.

compartment, where the radiopharmaceutical product is processed. Both compartments are accessible from the rear. While the biparting doors are open between the two compartments, neither the rear nor the front door to the entrance compartment can be opened; the operator is thus protected from possible exposure if he should inadvertently try to open one of these doors.

Of course, the construction cost of such an automated facility is relatively high; each cave, fully equipped, cost approximately $680 per square foot.

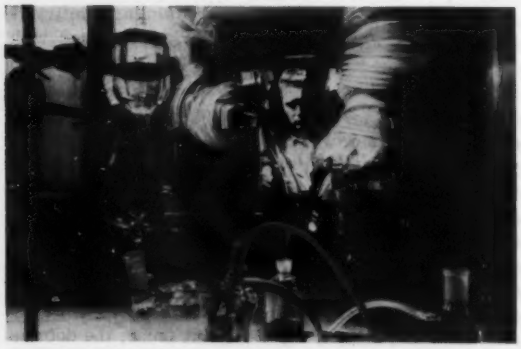


Fig. 3 Manipulators for remote operation inside the high-activity-level cave.

SPECIAL SEWAGE-DISPOSAL SYSTEM

To prevent contamination of public sewers with chance radioactive waste, two special underground storage tanks, 7500 gal capacity each, were provided. Waste water is collected and stored in these tanks until tests show that discharge to the main sewer is safe. The sampling is made through a permanent automatic system within the building.

Radiation Safety

A Radioisotope Committee is responsible for minimizing radiation exposure and ensuring the safe handling of radioisotopes. The Committee establishes regulations for the safe handling of these materials, and all their decisions are binding. They must approve all proposals for the use and distribution of licensed material within the plant, and they have the authority to interrupt work on any project where radiation is deemed excessive.

The Committee's regulations and decisions are implemented by the Radiation Safety Officer, who is responsible to the Committee. His duties are:

1. To witness all new or modified processes, establish safe radiation procedures, and spot-check occasionally to ensure that the operational monitoring is being carried out properly.

2. To monitor both restricted and unrestricted areas, which includes establishing survey and moni-

toring schedules, executing or supervising monitoring operations and evaluating the results, maintaining adequate records, and spot-checking.

3. To monitor personnel, evaluate and record both external exposure, e.g., by film badges and pocket dosimeters, and internal exposure, e.g., by thyroid checks and urine analyses, and to control skin and clothing contamination (Fig. 4).

4. To keep all legal records required by federal and state agencies as well as reports of radiological activities, e.g., daily tags, periodic summary reports, unusual incident reports, special work permits, minor injury reports and requests for medical service, records of receipt of radioactive materials and shipment, and records of all locations where radioisotopes are used at the facility.

Fig. 4 Hand and shoe monitor at which all persons leaving the manufacturing area are required to check out.

In addition, the Radiation Safety Officer must be thoroughly familiar with all state, federal, and management regulations pertaining to processing and handling of radioisotopes and radiation-producing devices; advise supervisors and personnel on the radiation-protection program; evaluate hazardous operating conditions and suggest to management safe procedures; establish emergency procedures for all dangerous operations; supervise decontamination after any dangerous spill; educate workers in the principles of radiation safety and in the hazards involved in working with ionizing radiation; and perform studies in the field of radiation protection.

Quality Control

The Quality Control Group checks all raw materials used, establishes standards for all packaging components, and rigorously tests finished products for both radioactivity and sterility prior to approval for sale. Product criterions are often more stringent than those set by the various federal agencies.

A unique feature of the Squibb facility is a machine developed by instrument companies, to our specifications, for counting and sorting the radioiodide capsules used for thyroid-function testing (Fig. 5). The percentage of iodine taken up by the thyroid is determined by administering to a patient a radioiodide capsule of known activity and comparing the activity over the thyroid area with that of an identical capsule. The filled capsules must be assayed and sorted into batches of identical activity, but manual counting and sorting are impractical for large-scale production.

The Squibb sorting machine picks a few capsules at random from the total batch, sufficient to provide a normal distribution curve. A capsule selected from the midpoint of this distribution is accurately assayed by an ionization-chamber method and designated the "standard capsule." The instrument is now

Fig. 5 Capsule-assaying device which automatically assays and sorts radioactive capsules into various activity groups.

calibrated so that a capsule whose activity is the same as that of the standard will be diverted to the 100% bin, which is the center bin of a total of 11. A capsule whose activity is 101% of the standard will be diverted to the bin immediately to the right of the 100% one, and a capsule whose activity is 99% of the standard will be diverted to the bin immediately to the left. The bins at the extremes collect capsules whose activities are greater than 104% or less than 96% of the standard, which are discarded. When the complete batch has been counted and sorted, a number of capsules from each bin, based on a statistical sampling plan, are randomly selected and manually counted as a final check.

The Future

The field of nuclear medicine is on the threshold of tremendous progress. To quote the Attorney General of the state of New Jersey, Arthur J. Sills, who, at the dedication ceremonies of the new Squibb radiopharmaceutical building, cited the Squibb Company for its leadership in nuclear medicine: "The availability of radioisotopes from this laboratory will bring new hope to many. The building is a monument to victories being won in the war against disease and to the contribution to health being made by this nation's pharmaceutical companies and their scientists." (MG)

Section II

Isotopes and Radiation Technology

Isotope Technology Development

Instrumentation for Snow Gaging—Yesterday, Today, and Tomorrow

By James L. Smith*

Abstract: Snow water content may be measured by any one of three basic systems: gravimetric sampler, pressure-pillow, and nuclear gage. Each of these systems is discussed and evaluated as to its usefulness. The development of each system is presented, and an attempt is made to predict the future instrumentation needed for study of snow structure and for measurement of snow water content for prediction of streamflow.

Most of the great river systems of the world have their beginnings in mountain snowpacks. Here precipitation in the form of snow collects during the winter, melts in the spring and early summer, and flows to arid lowlands where people, industry, and agriculture are usually found.

In addition to rainfall, the amount of snow that collects and the rapidity of melt determine the amount of water to be found at any one time in the streams, with too rapid a melt resulting in flooding. Since the early 1800's observations have been made of snow cover, and efforts have been made to relate cover and melt to streamflow. As our water needs have increased, dams have been constructed to store excess flows in spring to make water available for summer use. And as civilization encroaches on river valleys, flood damages increase. Therefore the need for predicting and controlling streamflow levels has become increasingly important.

The economy of much of the world depends upon a sustained, predictable amount of water originating from mountain snowpacks. It is imperative that we know the amount of snow and something of the melting rate to provide an orderly distribution to users, to plan for storage facilities, and to provide flood warnings sufficiently in advance of floods for people and their belongings to be moved from threatened areas.

In 1909 the system of snow surveying to determine the water content of a snowpack was developed. The methods most used have not changed since then, three basic forms of measurement having been used to date. The first, and most extensively used, method consists in one or more of the extraction or gravimetric techniques. The second, and currently popular, method makes use of a weighing system. The third, a recent innovation, uses isotope snow gages.

Gravimetric Sampler

The first report[1] of recorded snow measurements in the United States was made in 1836 by the Chief Engineer of the Chenango Canal, a Mr. Jervis, to the Canal Commissioners of the State of New York. He stated: "Snow on the ground which fell in November and December 1834 on the watershed of Madison Brook (9.37 sq miles) amounted to 87,120,000 ft^3 of water." We do not know how he measured the snow depth and water content. Still, his report indicates that a snow survey was made, and, as such, it is the first snow survey recorded in the United States and likely the first anywhere.

In a letter[2] dated Mar. 7, 1903, Charles A. Mixer, Resident Engineer of the Rumford Power Company in Maine, reported his method of snow measurement: "My usual method of gaging snow on the ground consists simply of getting a correct and full sample of the snow on the ground and then melting it to get the water equivalent. The sample is secured by forcing a cylinder down to the ground and then shoveling down around it and inserting a sheet metal bottom and lifting it out."

In 1895 a mountain-climbing young professor of Romance Languages, newly arrived at the University of Nevada from the University of Michigan, climbed to the top of Mt. Rose (10,800 ft elevation). This climb began the life work of Dr. James E. Church, the father of snow hydrology. In 1906 he offered to climb

*Project Leader, California Cooperative Snow Management Research Project, conducted by the Pacific Southwest Forest and Range Experiment Station, Forest Service, U. S. Department of Agriculture, in cooperation with the State of California Department of Water Resources and the Division of Isotopes Development, U. S. Atomic Energy Commission. See also J. L. Smith and D. W. Willen, Radio-Snow Gages: A Review of the Literature, *Isotopes and Radiation Technology*, 2: 41-49 (1964).

the mountain once each month for a year to obtain temperature readings if the Weather Bureau would furnish the thermometers. The Bureau agreed, and the Mt. Rose observatory was founded. During the 10 years that followed, instruments were installed to record air pressure, temperature, humidity, wind movement, precipitation, and sunshine. During this period Church developed many of his ideas of snow measurement and its relation to runoff.

EARLY SAMPLERS

In the winter of 1908–1909, Church developed a measuring device that became known as the Mt. Rose Snow Sampler and Scale.[3,4] This tool with some modifications is still the most commonly used tool for snow measurement. Church's new method consisted in forcing a 1.5-in.-diameter metal cylinder into the snow, withdrawing the sampler with included sample, and weighing the whole assembly on a special scale calibrated so that a 1-in. depth of water equaled a snow-core weight of 1.02220 oz (Fig. 1).

Fig. 1 Mt. Rose Snow Sampler with included core of snow being weighed with the Utah Sampler tubular scale.

With the development of a method for practical measurement of snow density, depth, and water content came the development of snow surveying and its use in flood and streamflow prediction. Snow surveying was begun at Mt. Rose, Nev., in 1909 by Church to answer the questions of how much water would flow from the surrounding mountains into Lake Tahoe and whether or not snowmelt water would cause flooding of the surrounding villages. These surveys were the beginning of a worldwide system from which flood and streamflow forecasts are made for much of the civilized world. In California alone more than 300 snow courses are measured at monthly intervals from January to June. From these measurements almost all the streamflow of the state is predicted.

In 1922 Korhoven[5] developed a sampler consisting of a 50-cm-high cylinder with a cross section of 100 cm^2. The cylinder was pushed into the snow and dug out with a shovel; the lid was then put under the sampler to hold the snow in the tube, and the whole assembly was weighed on a balance with a sliding weight.

Snow surveys were begun in Utah in 1923. An outgrowth of this work, the Utah Sampler, was developed and reported by Clyde[6] in 1932. It is similar to the Mt. Rose Sampler and has a throat diameter of 1.4872 in. so that 1 in. of water-equivalent of snow equals 1 oz. This diameter permits the use of ordinary commercial scales. A new scale was developed to go with the new sampler and has since become the standard scale for use with the Mt. Rose tube (Fig. 1). The scale is of the spring type and cannot be set to zero for empty-sampler weight. Therefore the weight of the empty tube is subtracted from the weight of the filled tube for each measurement. The Utah Sampler was designed with 12-ft sections instead of the 15-ft sections of the Mt. Rose Sampler.

From 1912 to 1965 the literature in snow hydrology is dominated by reports of gravimetric snow samplers of two basic types. Beginning with Mixer's[2] work in 1903 and continuing until 1954 we have a succession of investigators reporting on equipment for coring a large sample of snow, digging it out, and melting it to obtain the water content. Proponents of the Mt. Rose–Utah Sampler have been equally prolific, even to the present, in reporting various modifications of sampler design, section length, cutters, and material for tube construction.

In 1954 Zeleny Vaclav[7] of Czechoslovakia described a sampler 94.5 mm in diameter and 100 cm long whose sample was melted with a blowtorch. As recently as 1959 Sternzat and Sapozhnikov[8] of Russia described several instruments for measuring snow depth and density. Among these was a sampler very similar to the Mt. Rose, but extracted with a shovel. Density was measured with a portable beam balance.

MODERN SAMPLERS

In 1965 an exhaustive evaluation of the major Mt. Rose-type snow samplers was published.[9] The Federal Sampler (commonly referred to as the Mt. Rose Sampler) was found to overmeasure the water-equivalent of snow from an average of 7% in shallow,

light-density snow to 10–12% in deep, high-density snow. The overmeasurement appeared to be related to the shape, arrangement, and sharpness of the cutting teeth. Blunt-tooth cutters showed the greatest overmeasurement error, sharp-tooth cutters the least. The report suggested that a cutter might be developed which could reduce this overmeasurement and yet retain the ability of the sampler to cut and hold cores and still penetrate deep, dense snow.

The present Federal Sampler consists of a slotted aluminum cylinder with a cutter-point inner diameter of 1.485 in. Thus 1 in. of snow water in the tube weighs 1 oz. The tubular spring-actuated scale of the Utah Sampler is used for measurement.

SIPRE SAMPLER

The Snow, Ice, Permafrost Research Establishment (SIPRE) of the U. S. Army Corps of Engineers began snow-density determinations using small metal cylinders of 500 cm^3 volume to study internal snowpack densities. Its measuring device, known as the SIPRE Sampler, is still used in the standard method of internal-density measurement. A pit is dug in the snowpack, and SIPRE tubes are pressed into the face of the pit wall and then dug out and weighed to determine snow density and water content. Such samplers have an inherent error of about 5% (Fig. 2).

USE OF GROUND RADIOACTIVITY FOR SNOW MEASUREMENT

The use of the natural radioactivity of the soil is one of the newest experimental techniques for measuring the snow cover.[10] An area may be flown over prior to deposit of the snow cover and a plot of the natural radioactivity of the soil surface determined from recordings made by equipment in the plane. The radioactivity can be determined similarly after snowfall, when readings will, of course, be lower. The measurements obtained in the plane must be checked against snow-cover values determined from ground control stations. Theoretically the system appears promising.

Pressure-Pillow Method

Beaumont[11] suggested in 1957 the practicality of determining the total amount of snow overlying a point by weighing the snow. He proposed using a pillow as a weighing device. The pressure pillow is a butyl rubber pillow filled with methyl alcohol and installed on the ground prior to snowfall. As the snow falls on the pillow, the internal pressure is increased. The pressure is related to the mass, or weight, of the snow above it. A manometer or a pressure transducer may be used to measure this pressure. A common arrangement consists of monitoring weight changes on a stream gage recorder whose float rests in the manometer.

Beaumont reported that a 12-ft-diameter pressure pillow can provide an accurate measurement of the water content of the snow. The pillow is normally calibrated with measurements taken with the Mt. Rose Sampler. The output is a single value for the entire amount of water overlying the pillow, and response time after snowfall is a matter of minutes. However, rates of fall as low as 0.03 in./hr have been recorded, and pillows smaller than 12 ft in diameter did not provide accurate measurements. There are some minor problems involved in use of the pillow; for example, under some conditions it appears that snow bridging occurs. However, records obtained from it are normally accurate. The output may be plotted on an analog chart or telemetered to distant locations. The pillow has become widely used in the Western States. Its low installation and maintenance costs make it particularly desirable.

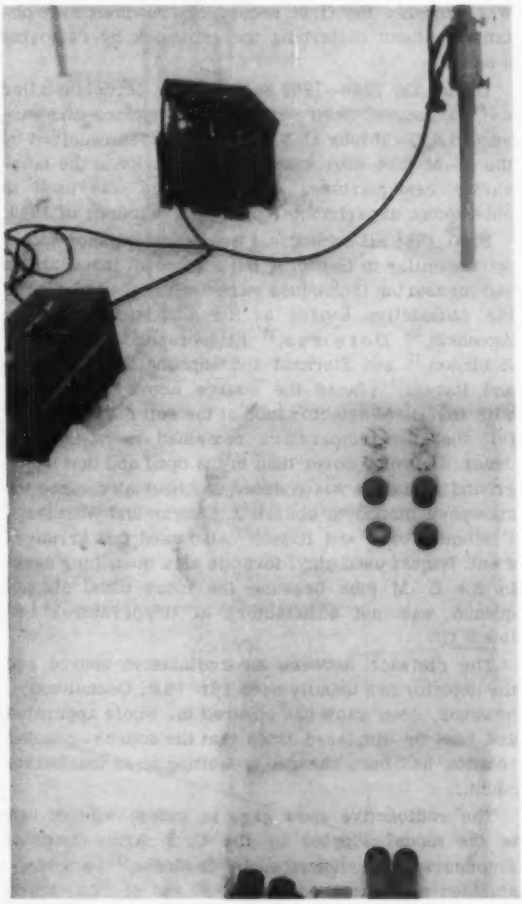


Fig. 2 SIPRE tubes used for measuring snow density. Note the single-access-tube gamma density gage and scaler.

Nuclear Gages

SINGLE-POINT FIXED-LOCATION GAGES

The most recent development in snow gages—the use of radioisotopes—was first reported by Gerdel, Hansen, and Cassidy[12] in 1950. The nuclear snow gage is designed to measure the water content of a snowpack lying between a radioactive source and a detector. This gage telemetered information to the Central Sierra Snow Laboratory (then under U. S. Army Corps of Engineers and now under the U. S. Forest Service) at Soda Springs, Calif.

The preliminary experiments of Gerdel et al. in the spring of 1948 had involved gamma emission from 20 mc of radiozinc installed beneath a 40-in. snowpack of 50% density, i.e., 20 in. of water. A G–M tube and a survey meter connected to a recording potentiometer produced a strip chart that recorded snow depletion for 1 week. Gerdel remarked that this was probably the first record of snowmelt ever obtained without disturbing the snowpack by removing a sample.

During the 1948–1949 snow season, Gerdel installed a ^{60}Co source flush with the soil surface and suspended a G–M tube 15 ft above it. Pulses received by the G–M tube were transmitted by radio to the laboratory headquarters. A ^{90}Sr source was used in subsequent experiments during the summer of 1949.

Until 1965 all subsequent field-location snow gages were similar to Gerdel's, but equipment installations and measuring techniques varied. Gerdel et al. placed the radioactive source at the soil surface, as did Agashkin,[13] Doremus,[14] Hildebrand,[15] Martinec,[16] Robinson,[17] and Sternzat and Sapozhnikov.[18] Higashi and Itagaki[19] placed the source above the ground, with the G–M detector tube at the soil surface. They felt that the temperature remained more constant under the snow cover than in the open and that background radiation was reduced or eliminated since the snowpack tended to absorb it. Duncan and Warnick,[20] Fischmeister,[21] and Itagaki[22] also used this arrangement. Itagaki used ethyl formate as a quenching agent in the G–M tube because the more usual alcohol quench was not satisfactory at temperatures below 0°C.

The distance between the radioactive source and the detector has usually been 10 to 15 ft. Occasionally, however, deep snow has covered the whole apparatus and bent or displaced it so that the source–counter relation has been changed, resulting in an inaccurate count.

The radioactive snow gage in widest current use is the model adopted by the U. S. Army Corps of Engineers. It is described by Robinson[17] as a lead-shielded collimator containing 80 mc of ^{60}Co, which is buried in the ground; a scintillation detector is mounted on an H-frame structure 15 ft above the collimator. The detector is a 1.5-in.-diameter thallium-activated NaI crystal with a photomultiplier tube. Shims are mounted over the collimator to compensate for source decay. The system is designed for use with a maximum snow depth of 50 in. water-equivalent.

Various depths of water-equivalent measured by the radioactive-snow-gage method have been given by other investigators;[13–22] they range from 2 to 52 in. Beyond this depth the absorption of radioactivity by the snow is so great that transmission of gamma radiation from the source to the detector is drastically interfered with.

The readout from the single-point fixed system is a single number translatable to the water-equivalent of the entire snowpack lying between the detector and source. The condition of the pack, such as its density, ripeness, and included ice layers, cannot be detected nor can changes within the pack due to heat or rain input. Therefore the depth–density changes in the pack with time cannot be determined. But single-point fixed gages can be telemetered. They give data comparable to those obtained with the Mt. Rose Sampler and with the pressure pillow.

AIRBORNE RADIOMETRIC SNOW GAGE

An airborne radiometric snow survey system has recently been developed by the Electronics Research Laboratory of Montana State University[23] under contract to the Bonneville Power Administration of the U. S. Department of Interior.

In remote locations where access is difficult, an airborne system presents several advantages. It does not require the landing of aircraft under hazardous conditions and is not affected by failure of telemetry equipment or of on-site, hard-to-reach electronic equipment. In the airborne system a radioactive source, 20 curies of ^{60}Co, is buried in the soil prior to the snow season and shielded from the side by a lead collimator. A cone of radiation is sent skyward. A plane carrying electronic detection equipment is flown through the radiation beam at a preset altitude between 100 and 300 ft. Detection equipment in the plane consists of a scintillation detector, pulse-height analyzer, scaler rate meter, and digital readout. This system is controlled by electronics recording the amount of time spent in the cone of influence. In tests made over two winters, errors between values recorded in successive flights on the same day averaged 3 to 5% from the mean. Comparisons between the water content found by the Mt. Rose Sampler and that found by the airborne method showed variations of as much as 7% between the two. The error was greatest when the water content of the pack was 4 in. or less.

PORTABLE SINGLE-PROBE SCATTER GAGES

Single-access-tube neutron soil-moisture and gamma soil-density gages have been used to measure

snow water content and density.[24] These units emit nuclear energy—neutrons in one case and gamma rays in the other—which reacts with the surrounding medium and is scattered back to the detector. They have a sphere of influence of from 9 to 16 in. diameter (Fig. 3), and thin layers whose densities differ from the average surrounding density are difficult to measure. In addition, there is a large energy loss in the 12 in. nearest the snow–air and snow–soil interfaces. Such systems require three or four calibration curves. Smith and Willen[25] reported measurement of total snowpack water-equivalent to within ±2% accuracy with both neutron and gamma single-access-tube systems when gravimetrically calibrated against a sphere-of-influence sized sample of snow. When the ^{137}Cs source from the gamma system was removed from the detector probe and dropped into another access tube 12 in. distant from the detector probe, it was possible to measure snow density to 98% accuracy in 3-in. vertical increments. To make such measurements, however, four separate calibration curves were required because of the energy loss at the air–snow interface (Fig. 4).

TWIN-PROBE GAMMA-TRANSMISSION SNOW GAGE

Since the extraction or gravimetric method destroys the sample site, it is unsuitable for precise work if the same site must be sampled repeatedly. The single-point fixed-location snow gage of the Gerdel type yields total water content only. The portable backscatter neutron and density probes are unsuited for density measurements in the important surface 12 in. and cannot be used to measure narrow

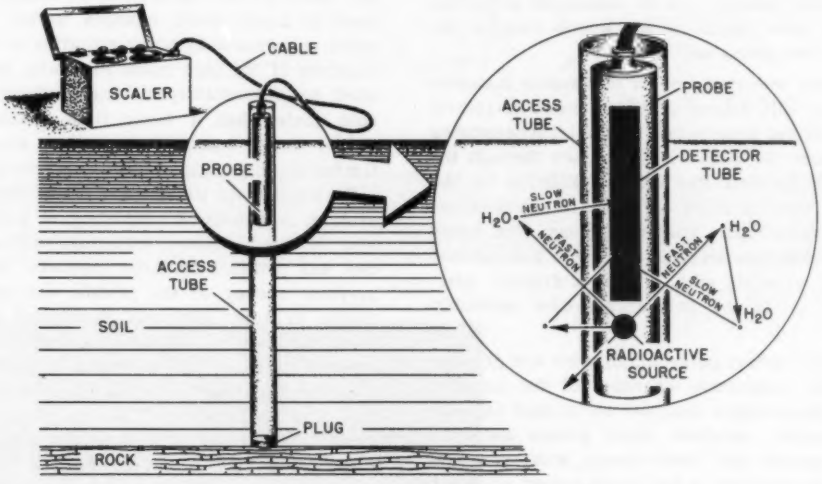


Fig. 3 Nuclear-Chicago neutron soil-moisture meter installation with spherical volume of influence. (Use of trade name is for information only and does not constitute endorsement by the U. S. Department of Agriculture.)

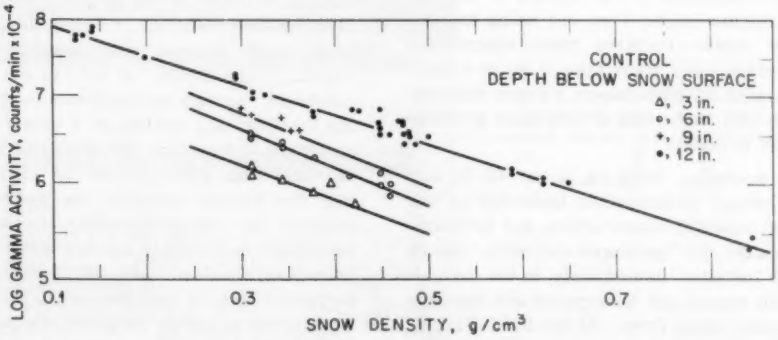


Fig. 4 Control calibration curves for the gamma-transmission probe.

snow bands whose density differs from that of the general pack except as separated source–detector systems, and in this case the work is very slow. The pressure pillow, while giving accurate measurement of total water content, cannot be used to study internal structure.

At the Central Sierra Snow Laboratory, a portable gamma-transmission profiling twin-probe system of snow-density measurement has been developed with which the total water content of snowpacks, their density in 0.5-in. vertical increments, and their in situ density changes with time may be measured.[26] The system may be operated either manually or remotely with telemetering of its data. Since one may measure pack density changes within a single profile as often as desired with this system and can concurrently study causative factors, it is now possible to relate causative factors to snowpack density changes. Since density can be measured in 0.5-in. increments, such items as ice lenses may be detected within the pack itself.

This system was designed to accurately measure the mass per unit volume of 0.5-in.-thick horizontal layers of material lying between parallel access tubes extending from the snow–air interface through the pack and into the soil. The source, sealed in the end of a holder rod, is 5 mc of ^{137}Cs and provides essentially monoenergetic gamma photons. The probe unit is a scintillation detector [0.5- by 1.5-in.-diameter NaI(Tl) crystal] with a photomultiplier tube. The source is in the twin-access-tube assembly (Fig. 5).

The detector output-pulse amplitudes are proportional to the respective energies of the incident photons. A pulse-height analyzer set to pass only the energies desired receives these pulses and discriminates against any lower energy, scattered photons. When the window in the pulse-height analyzer is set to pass only the energy of the ^{137}Cs photopeak, only those unscattered photons are counted which have traveled, without collision, in a straight line from the source to the detector crystal. The number of photons is proportional to the density of the material in the volume (about 4 cu in.) being studied. A scaler rate meter receives these unscattered pulses and electronically translates them to counts, which are indicated on glow tubes. A chart recorder attached to the rate meter via an amplifier provides an analog record of counts.

The unit is portable, weighing about 60 lb, and operates from either rechargeable batteries or line voltage. It is of modular construction, and downtime is minimal because the "packaged circuitry" can be substituted and installed immediately in the field. It is electronically stable and waterproof and operates over a temperature range from −23 to +60°C (Fig. 6). The calibration curve (Fig. 7) of the system follows a simple linear log regression relation, and, when used with care, the system is accurate to within ±1.2% of the actual snow density.

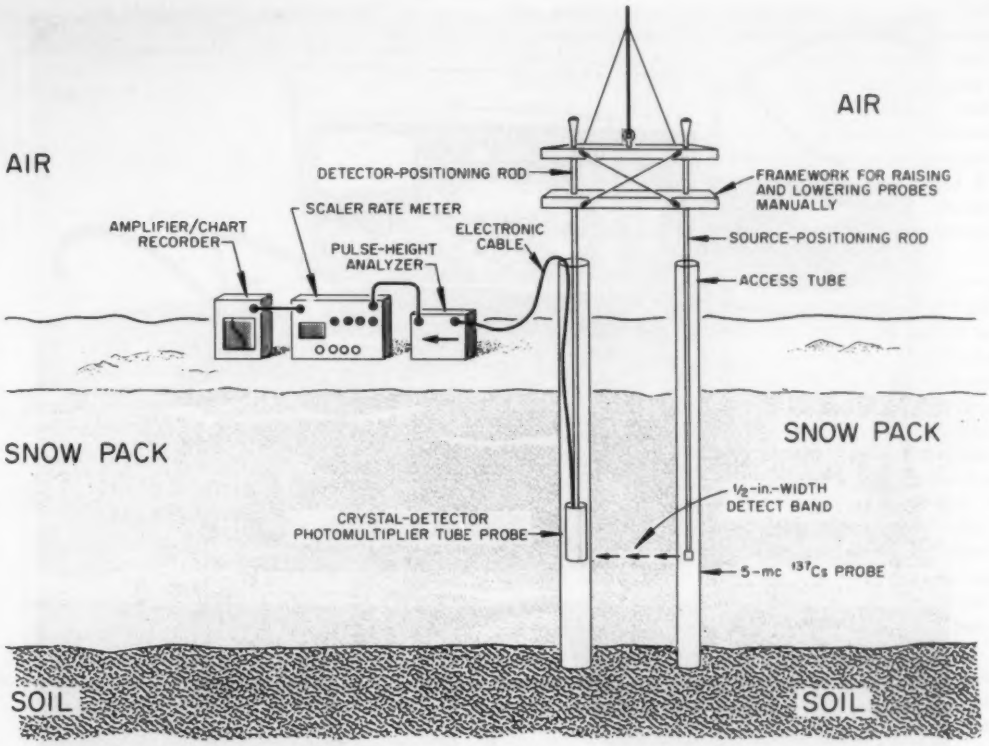
To test the ability of the system to detect density changes, a natural snowpack was partly replaced with ice lenses of known density. Access tubes were installed, and the source and detector were simultaneously lowered to counting depths. Zones of density change were located by first scanning the entire profile with the rate meter. One-minute counts were then made at 1-in. intervals throughout the profile except near and across zones of density changes. At such points counts were made at 0.5-in. profile intervals (Fig. 8).

With the system it is possible to delineate different density zones within a snow profile to 0.5-in. vertical increments. Thus the gage can be effectively operated at any point from 0.5 in. below the snow–air interface to within 0.5 in. of the snow–soil interface and within the soil underlying the snow. It can even be used to index small changes in the snow surface which are caused by solar radiation or other stimuli. Because of the high count recovery, the gage can be used as a constantly moving profiling system. The rate meter has a 4-sec time constant, which is sufficient to smooth out random statistical count fluctuations inherent in radioactive decay.

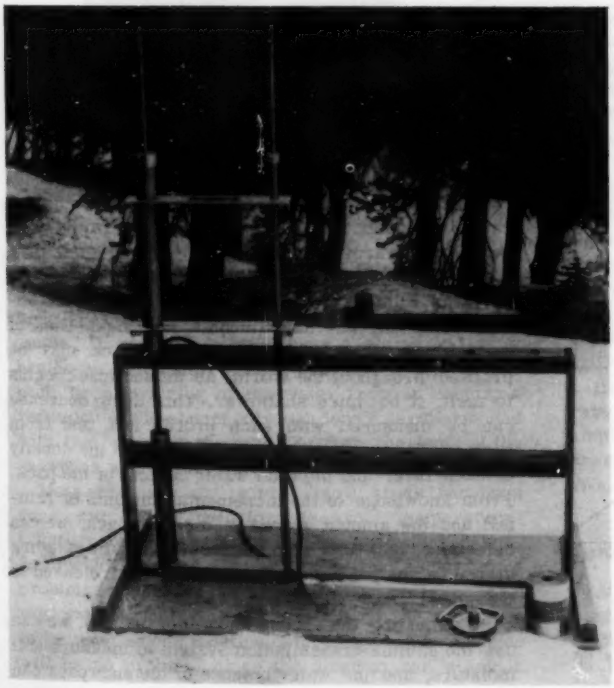
To provide an illustration of the sensitivity of the system, a snowpack consisting of a natural profile with included lenses of materials with varying densities was profiled by three methods. First, a point-by-point survey of the profile was made by taking counts at 1-in. intervals throughout the snowpack. Then the source and detector were simultaneously lowered through the snowpack at speeds of 10 and 5 sec/in. Reproduction of the actual snowpack density was most accurate at 5 sec/in. Repeat runs at the same speeds resulted in curves of almost exactly the same magnitude in every portion of the curves, although the source and detector were lowered by hand and a stopwatch was used. Scans at a rate of 5 sec/in. reproduced the pack conditions more faithfully than did the more laborious point sampling.

One of the most important applications of this equipment could be as an early-warning flood-forecast system. Detection of water movement in a snowpack could indicate the beginning and ending of rainfall. The ability of the system to detect water movement through the pack was determined by placing the detector and source at a common depth within a snowpack and plotting the change in density with time as water was sprayed onto the snow surface. In this test the system detected the wetting front when it reached the source-detector horizon. The density increased in response to new water in the pack and remained constant until the addition of water was stopped. The pack then drained to its original density. The system faithfully recorded the pack performance.

This snow gage may be used as a portable or an automated, telemetered high-altitude monitor, or



(a)



(b)

Fig. 5 Portable collimated gamma-transmission snow-density gage. Access tubes may or may not extend into the soil. (a) Diagram; (b) in place.

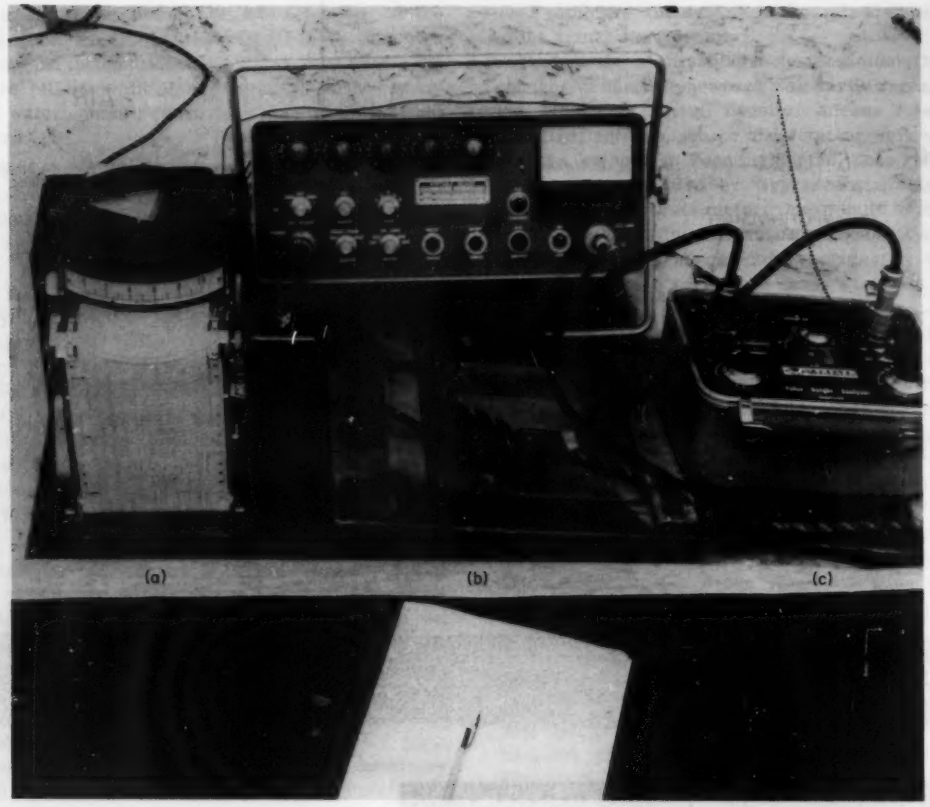


Fig. 6 Electronic system for snow-density gage shown in Fig. 5. (a) Troxler Laboratories SC-10 Density System, (b) pulse-height analyzer, (c) scaler rate meter and amplifier–chart recorder. (Use of trade name is for information only and does not constitute endorsement by the U. S. Department of Agriculture.)

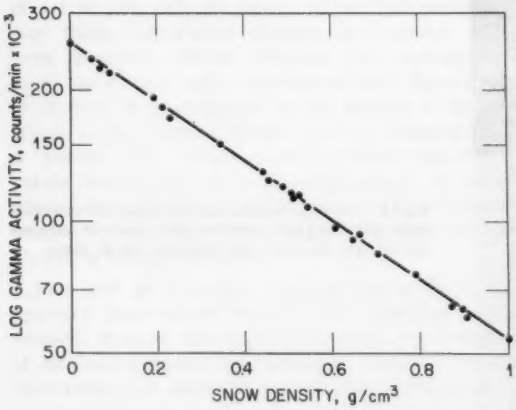


Fig. 7 Calibration curve, which is accurate for all densities and for all depths from 0.5 in. below snow surface up to within 0.5 in. of the underlying soil.

"snow sentinel," of the snowpack performance during rapid snowmelt. Melt may occur from either unseasonably warm weather or from rain on snow. In a rain-on-snow situation the density of the various horizons within a snowpack may be determined before a storm, as may amounts and intensities of rainfall during a storm since the pack may be profiled throughout the storm. As the snowpack begins to melt, it becomes shallower. This depth decrease can be measured with each profile run, and from each profile run we may also determine the density of each layer and the total water content of the pack. From knowledge of the incremental amounts of rainfall and the amount stored in the snowpack, we can determine how much of the incoming rain is being stored in the pack and how much is being released to the soil or as overland flow.

By extending the access tubes into the soil, we can use the gamma-transmission system to measure soil moisture, and the water balance of the snowpack can

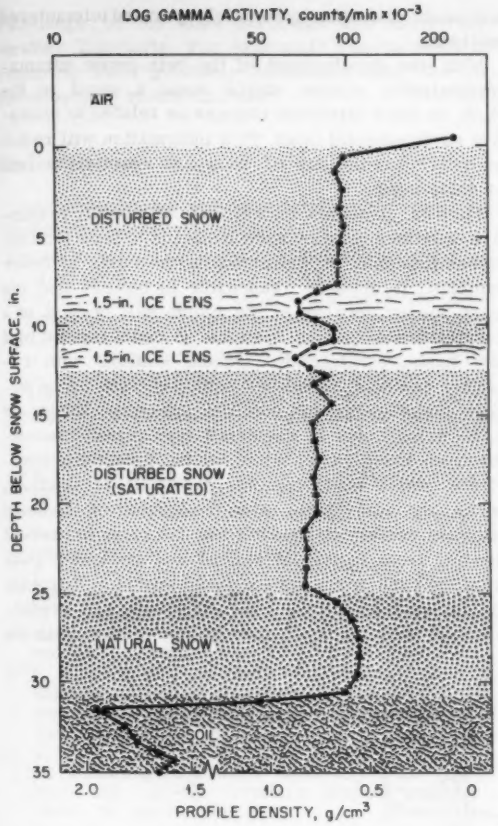


Fig. 8 Density profile of snowpack.

be extended to take into account the water lost to the soil.

The gage will prove useful in studying the effect of rain on snow. Storms of various types may be artificially imposed upon a snowpack and the pack density profile monitored. This information will provide the basis for prediction of snowpack performance under various storm situations. For example, if rainfall stops during a storm and snow begins to fall, this change will be reflected in the pack by a draining of the water from it, causing a reduction of the abnormally high densities experienced with free water in the pack. Further, the depth of the snowpack will begin to increase.

Most research on isotope snow gages has been done with G–M detectors to avoid temperature-caused problems inherent in a scintillation system. Smith, Willen, and Owens[24] found that temperature problems could be minimized to permit accurate work with the scintillation system. If the system is to be used in one location only, temperature changes will be no problem since the detector can be left in the snowpack at all times. However, if the system is used at many locations in a single day, temperature changes will pose a problem. In such a case the detector will heat up while outside the snowpack and cool down inside the pack. This problem can be minimized by permanently insulating the detector probe. Further protection can be provided by carrying the detector probe from location to location in an insulated carrying case. Use of a peak-stabilized pulse-height analyzer to provide automatic adjustment of the window should eliminate this problem.

One of the most-often-cited objections to snow gages that use access tubes is the problem of snowmelt around the tube, mainly in the surface 12 in. of the snowpack. This so-called "suncupping," which is caused by radiation of heat from the tube to the snowpack, causes an air gap to form and thus decreases the amount of snow between the detector and source. If the air gap is large enough it can affect the accuracy of the density measurements, which could be particularly troublesome if the access tubes were spaced only 12 in. apart. Suncupping is usually not large enough with any type of tubing to affect the count accuracy if the tubes are spaced 24 in. apart. The best form of access tubing is aluminum for strength, surrounded by about 0.25 in. of white expanded polystyrene for insulation.

Because of the high resolution of the detector in the scintillation system and the low density of the snow, access tubes may be spaced 2 ft apart without any loss of accuracy or significant loss of operating speed and still use a 5-mc ^{137}Cs source.

During the winter of 1965–1966, a system for semiautomatic operation of the gamma-transmission profiling snow gage was developed and installed at the Central Sierra Snow Laboratory (Fig. 9). The probes were suspended in their respective access tubes from a boom projecting from a nearby tower and were raised and lowered by a cable from the boom. The electric line to the winch and the line to the detector probe were run from a nearby equipment shelter, which housed the pulse-height analyzer, rate meter, and amplifier–chart recorder. To profile a snowpack it was only necessary to turn on the electric motor driving the winch, the rate meter, and the chart recorder. The rest of the operation was automatic. The probes rose automatically at a rate of 1 ft/min (5 sec/in.), the millivolt output from the rate meter was changed to milliamperes in the amplifier and automatically plotted on the analog chart recorder, and the recorder charts were quickly reduced to digital form in the office.

Future of Snow Gaging

At the Central Sierra Snow Laboratory, we are designing the profiling snow gage for telemetered remote operation. In this system the probes would be moved from below ground in sealed tubes. At-

tached to endless perforated steel tapes riding on pulleys, the probes would be driven from below ground by a small electric motor. Current to operate the motor would be provided by rechargeable batteries whose voltage would be maintained by a wind-driven or thermoelectric generator. The electronic components would be essentially those of the present gamma-transmission profiling snow gage with some modification. The source would be 5 mc of ^{137}Cs and the detector a 1.5- by 0.5-in.-thick NaI(Tl) crystal and photomultiplier. The signal would pass to a peak-stabilized pulse-height analyzer designed for cold-weather operation on 12-volt direct current. From the pulse-height analyzer the signal would go to a rate meter, then to a digitizer, and finally to a data phone set for transmission either over a phone line or by radio signals. The telemetry package would allow for actuation of and shut down of the gage from distant locations.

Within a short time orbiting communication satellites will be used by hydrologic networks as radio relay stations. Such an advancement could bring an end to problems of transmission from mountainous country. And the mounting increase in labor costs makes it obvious that there will be a decrease in manual collection of snow data and an increase in automatic collection by remotely operated telemetered stations.

With the development of the twin-probe gamma-transmission system should come a spurt in the study of snow structure changes as related to causative environmental input. This information will result in better predictions of snowpack reaction to rain or solar input.

Further, it seems logical that, with better predictive equations relating snow behavior to input factors, streamflow may be predicted on the basis of fewer samples. It appears reasonable to believe that the snow-survey network of the future will consist of a combination of gaging systems. It could consist of a few basic, strategically located stations with remotely operated, telemeter-equipped, twin-probe nuclear snow gages and satellite pressure pillows or U. S. Army Corps of Engineers-type gages. The output from these gages would be fed directly to computers, which would compute snowmelt, streamflow, peaks, and other data—even the number of sandbags and men needed at bend "X" in the river to prevent flooding from individual storms and snowmelt. Data from the twin-probe system would precisely delineate the internal condition of the snowpack, and predictions as to melt could then be made from intimate

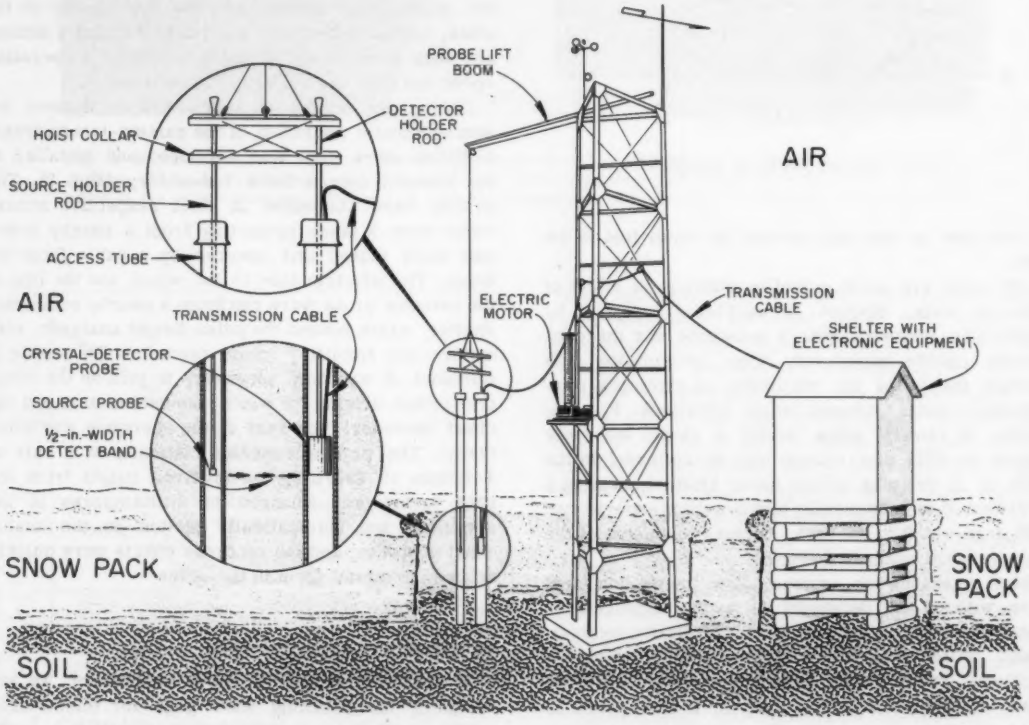


Fig. 9 Elements of the permanently emplaced snow gage.

knowledge of the snow structure and short-range weather forecasts. For extremely remote sites not requiring such precisely delineated data, the airborne system should find use. (MG)

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Oxygen Determination by Nuclear Methods

By F. J. Miller

Abstract: Oxygen determination by activation analysis and other nuclear methods has been shown to be feasible for many different materials. The development of the compact 14-Mev neutron generator and its commercial availability have made its use for oxygen determination very nearly competitive with conventional methods in specific applications. The rapidity of analysis and ease of operation of the 14-Mev generator technique are features of chief interest to the steel industry. Other radiochemical methods of analysis are uniquely suited to applications such as the determination of oxygen in oxide films, analysis for tracer ^{18}O, and the indirect determination of oxygen in water.

Oxygen is an element of concern to widely different disciplines. Metallurgists, studying the effect of oxygen on the physical properties of different metals, have become increasingly interested in the presence of part per million amounts of oxygen in metals—both as dissolved oxygen and as oxides. Geologists, seeking to classify rocks, need to know the oxygen content as an aid to identification. Biochemists, recognizing the intimate relation of oxygen to life pro-

cesses, require a means not only of ascertaining the quantity of oxygen present in a living system, but also of following the oxygen through various metabolic pathways. Pressure from the needs of the many fields of interest has led to the development of methods for oxygen determination that are rapid, precise, and sensitive. Nuclear activation analysis is assuming an important place among these methods. Tracer studies with ^{18}O and radio-release methods also may be more expedient and elegant in application than older methods of analysis.

In the following brief survey, the commonly used chemical methods are discussed for comparison with nuclear methods. The adaptability of nuclear methods for the determination of oxygen both in the chemically bound and free states for the many areas of interest is shown by selected references to work carried on in metallurgy, analysis of meteorites and lunar rocks, organic materials, biochemistry, and the newly burgeoning oceanographic sciences.

Activation Analysis

DETERMINATION OF OXYGEN IN METAL OXIDES

The determination of oxygen in oxides is always complicated by the abundance and pervasiveness of the element. Whether the element is determined by conventional methods or by nuclear methods, the success of the determination depends to a degree upon the handling, pretreatment, and homogeneity of the sample. If the determination of inclusions of oxide within a test specimen is desired, the surface must be cleaned, oxide layers removed, and any further accumulation of oxide or moisture on the surface prevented. Reduction of the size of the sample by grinding or slicing should be done under an inert, dry atmosphere.

Conventional methods of analysis of oxides for oxygen content are based on either carbon reduction at elevated temperature or replacement of the oxygen by fluorine. In carbon reduction the metallic cation is reduced either to the metal or to a carbide, and the oxygen is liberated as carbon monoxide. The carbon reduction can be performed as either a vacuum-[1] or an inert-gas-fusion[2] procedure. In both cases the test specimen is dropped into a carbon crucible that may also contain some platinum metal as a molten bath to aid in maintaining contact of the test material with the carbon surface. The carbon crucible, contained within a silica envelope, is heated, usually by a radio-frequency induction furnace, to a temperature sufficient to initiate the reduction reaction, 1800 to 2300°C.

The primary difference between vacuum fusion and inert-gas fusion is in the handling of the produced gas. In vacuum fusion the silica envelope is evacuated prior to reduction of the oxide. The gases that result from the reaction are pumped into a suitable measuring device. In inert-gas fusion a stream of inert gas flows continuously, sweeping the gaseous products into a measuring apparatus such as a gas chromatograph, a conductivity cell, or a manometer. Where weighable amounts of CO are produced from the sample, they can be converted to CO_2, collected on a solid absorbent, and weighed. The vacuum-fusion and the inert-gas-fusion methods have been highly developed by commercial manufacturers. Complete assemblies are available. An analysis may be completed in 6 min on standard inert-gas-fusion equipment. The vacuum-fusion procedures take slightly longer because of the pump-down time but are equally routine.

Despite difficulties in safe handling, the fluorination procedure, in which the oxygen present as an oxide is replaced with fluorine, is sometimes preferred. The oxygen may be liberated as oxygen if $KBrF_4$ is used as the fluorinating agent[3] or as water if HF is the fluorinating agent. In either case the liberated gas is swept from the reaction chamber through appropriate traps to separate the undesired portions of the gas stream. The oxygen content is usually measured manometrically. If water is generated, the quantity may be determined by passage through an electrolytic cell. Some fluorination procedures entail the use of unstable compounds, e.g., $KBrF_4$, under pressure at moderately elevated temperatures. The compounds may react explosively, and the resultant products are toxic. Thus the fluorination reaction to release bound oxygen is used only when other methods cannot be used. The technique is particularly applicable in the case of fluoride salts that contain trace quantities of oxides.

Recently, activation methods for oxygen determination have been developed which will not supersede conventional methods but which can favorably compete with them in some cases. The 14-Mev neutron generator activation method can be invaluable, particularly in the case of a large steel mill or other metal refining plant where numerous oxide determinations are necessary. The rapidity and ease of analysis coupled with the economy of operation can offset the larger initial cost of the 14-Mev generator.

One commercially available 14-Mev neutron generator[4] uses a Cockcroft–Walton type accelerator that operates at 200-kv accelerating voltage with a beam current of up to 2 ma. The $^3H(d,n)^4He$ reaction is used to obtain 1 to 2×10^{11} n/sec at an energy of 14.2 Mev.

In the complete system a pneumatic transfer device moves the samples in polyethylene bottles into the irradiation position. After irradiation the sample is "blown" to a counting station, the count taken, and the sample transferred to storage. A 5- by 5-in. NaI(Tl) detector, a single-channel analyzer, and two scalers serve as counting equipment. A Lucite reference sample is used as a flux monitor for this system. The use of such a reference flux monitor minimizes errors due to space and time variations of the

neutron flux. The analyzer is set to accept gamma radiation between 4.6 and 7.6 Mev.

The 14-Mev generator activation method of oxygen determination relies upon the $^{16}O(n,p)$ nuclear reaction which yields ^{16}N, with a 7.35-sec half-life. According to Guinn,[5] oxygen levels as low as 5 ppm (in 20-g samples) can be determined rapidly and nondestructively by this method. Total time for the analysis is only about 1 min per sample.

In a comparison[4] of the analysis of steel samples (7 to 10 g, 1.5 in. long) by activation analysis, vacuum-fusion analysis, and inert-gas-fusion analysis, the difference in the results obtained by neutron activation and by vacuum fusion in 65% of the determinations was $\leq 0.001\%$ O_2; the correlation between neutron activation and inert-gas fusion was even better. The study indicated that the neutron activation analysis technique is at least as reliable as conventional techniques. Large samples (7 to 50 g) can be analyzed in half the time needed for smaller (1 to 3 g) samples by other techniques. Since the integrity of the sample is preserved, any questionable result can be verified by retrieving the sample from storage and redetermining the oxygen content. The use of larger samples minimizes the errors that may occur because of inhomogeneities in the steel. Other sources of error in the hot-reduction techniques, such as incomplete decomposition of the oxides, mechanical ejection from the carbon crucible, and the gettering effect of the recombination of the reduced metal with the released oxygen, are not present in the neutron activation method.

Interferences from other elements that may be present in the steel samples are few. Fluorine interferes because of the $^{19}F(n,\alpha)^{16}N$ reaction which yields the same isotope as the $^{16}O(n,p)^{16}N$ reaction. Interferences from the $^{27}Al(n,\alpha)^{24}Na$ and the $^{56}Fe(n,p)^{56}Mn$ sum peaks are minimized by setting the analyzer to accept gammas only in the range from 4.6 to 7.6 Mev. Boron can interfere because the $^{11}B(n,p)^{11}Be$ reaction produces radiation that can completely obscure the ^{16}N produced by the oxygen. Special precautions must be observed when the presence of fluorine or boron in significant quantities is suspected in the material to be analyzed.

In 1965 a cost comparison made at the Allegheny Ludlum Steel Corp. by Fricioni[6] showed that vacuum-fusion equipment cost \$10,000 to \$30,000 and required 10 to 12 min per oxygen determination; inert-gas-fusion equipment cost about \$9000 and took 6 min. Basic instrumentation for neutron activation cost about \$22,500. Shielding adds \$3000 to \$5000, and computer calculation with readout, approximately \$6000. But neutron activation analysis requires only 1 min exclusive of sample preparation time. Since this comparison was made, the cost of the basic neutron generator equipment has been reduced somewhat; however, the comparison is still valid. Evidently a large volume of samples with an extreme requirement for speed of analysis is needed to justify the cost of a neutron activation analysis system.

One problem encountered in the analysis of metals is the oxygen content of the container and of the entrained air, which can produce a blank value running as high as 100 ppm oxygen. Various means of coping with this problem have been attempted. Steele[7] encapsulated his samples in an inert atmosphere in copper tubing that contained only 5 ppm oxygen. Coleman[8] used a polystyrene capsule, long enough that one end remained unirradiated, and slid the sample into the unirradiated portion for counting. Other suggested alternatives include perforating the capsule to blow the irradiated air from it after irradiation, and removal of the sample by hand from the capsule prior to counting. One suggested technique[9] uses especially purified argon to move a bare metal slug into the irradiation position and thence to the counter so that the background value of the container together with that of the entrained air can be eliminated.

The 14-Mev neutron generator activation method is useful for the determination of oxygen in metals other than steel. Oxygen has been determined in Be, Mo, V, Ce, La, Cr, Sn, and W (Ref. 10). Anders and Briden[11] have reported a complete study of the determination of trace oxygen in cesium metal. They concluded that trace amounts of oxygen can be determined in very reactive and hard-to-handle samples by the neutron activation method. Byrne, Illsley, and Price[12] used the method to determine oxygen in beryllium metal. They found that, at an oxygen concentration of 0 to 2%, the relative standard deviation of their determinations was <4%, which compared favorably with results obtained by existing chemical methods. In addition, automatic activation analysis was nondestructive, much more rapid, and not subject to errors from oxidation of the sample chips. Errors could still result from possible nonhomogeneous distribution of oxygen in the test specimen.

The analysis of alkali metals other than cesium is also feasible by the neutron activation method. Steele[13] reported the determination of oxygen in potassium metal. He estimated possible error by determining an organic acid, that contained in the range of 10 to 100 mg oxygen, with a standard deviation of only 2%. He also discussed the improved detection equipment and sample handling equipment that led to the better precision and sensitivity.

Advantage has been taken of the nondestructive feature of the neutron activation method to analyze scarce materials, such as meteorites, for oxygen content. Vogt and Ehmann[14] analyzed stony meteorites satisfactorily, after which the entire amount of the test specimen remained available for chemical or other analysis.

REMOTE ANALYSIS

One of the more intriguing applications of the fast-neutron activation technique was proposed by Caldwell

et al.[15] for the remote analysis of lunar and planetary surfaces. The equipment required consists of a scintillation crystal detector, an accelerator neutron source of a miniaturized and pulsed design, a multichannel analyzer with pulse-height and time-analysis modes, and a programing device to control the overall experiment. The equipment would enable the analysis of lunar rocks for oxygen as well as some ten other elements. A knowledge of the ratio of the concentration of oxygen to iron would permit the classification of the rock into acidic, basaltic, or aerolytic rocks.

OXYGEN IN ORGANIC MATERIAL

The current widespread use of the neutron activation method to determine the oxygen content of metals and other inorganic substances should not obscure its applicability to organic materials. Stallwood et al.[16] studied the determination of total oxygen content of organic materials by fast-neutron activation. They concluded that the fast-neutron activation method allows oxygen to be measured faster and with better precision and accuracy than by any other method. Oxygen concentrations greater than 100 ppm could be routinely measured within 10% in <10 min; for 30 ppm, the relative standard deviation was ~25%. The oxygen content of the sample container material was the deterring factor in lowering the detection limit and improving the precision.

Other Nuclear Methods

The above discussion has been confined to the use of the 14-Mev neutron generator because of its growing use in industry. Other nuclear reactions may also be useful for oxygen determinations in specific instances. Bate[17] has tabulated a number of such reactions as shown in Table 1.

In his discussion, Bate indicates that the use of tritons has been found of value. The reactions involved are $^{6}Li(n,\alpha)^{3}H$ and $^{16}O(t,n)^{18}F$, followed by the decay of the 112-min ^{18}F by positron emission to ^{18}O. The reaction is initiated by reactor neutrons. This method requires the intimate mixing of a lithium compound with the sample. Lithium fluoride is useful for such purposes since it can be used as a carrier for the ^{18}F produced and for the $^{19}F(n,\gamma)^{20}F$ product. Interference from ^{20}F is minimal since ^{20}F has a half-life of 10.7 sec, which is short as compared with the 112-min ^{18}F, the isotope of interest. Other advantages of this salt over other possible salts are a lesser hygroscopicity, equal ratio of lithium to fluorine atoms, and a relatively low oxygen content. The reaction has been used for the analysis of beryllium metal powder.[18] Bate and Winchester, as cited by Bate and Leddicotte,[19] also used the method to determine oxygen in oxide films.

The thickness of the oxide film on tantalum foil was determined by Thompson[20] using protons of 4 Mev energy and a 10-μa beam current. The reaction is $^{18}O(p,n)^{18}F$. The positron annihilation radiation (0.51 Mev) from the decay of 112-min ^{18}F was counted. The maximum film thickness for the analysis with 4-Mev protons was estimated at 25 μ.

De Goeij and Houtman[21] examined further the use of the $^{16}O(t,n)^{18}F$ reaction originally proposed by Smales[22] for oxygen determination. Of particular interest to de Goeij was the limited range of the triton particle used to estimate the oxide concentrations in surface layers. An application of this type can be invaluable in metallurgy. Specific examples discussed were the determination of the thickness of Al_2O_3 layers on aluminum, determination of oxygen in Li–Al–Mg alloy, and the determination of surface oxygen in polyolefins.

Table 1 NUCLEAR REACTIONS OF OXYGEN

Reaction	Half-life of induced nuclide
$^{16}O(n,p)^{16}N$	7.35 sec
$^{16}O(t,n)^{18}F$	112 min
$^{16}O(\gamma,n)^{15}O$	124 sec
$^{16}O(t,p)^{18}O$	Stable*
$^{16}O(^{3}He,p)^{18}F$	112 min
$^{16}O(n,\alpha)^{13}C$	Stable†
$^{16}O(^{6}Li,n)^{21}Na$	23 sec
$^{16}O(^{6}Li,^{4}He)^{18}F$	112 min
$^{16}O(n,2n)^{15}O$	124 sec
$^{16}O(d,n)^{17}F$	66 sec
$^{16}O(^{3}He,n)^{18}Ne$	1.46 sec
$^{16}O(d,p)^{17}O$	Stable‡
$^{16}O(p,n)^{16}F$	Short
$^{16}O(\alpha,n)^{19}Ne$	18 sec
$^{16}O(d,\gamma)^{18}F$	112 min
$^{16}O(\alpha,pn)^{18}F$	112 min
$^{16}O(p,\alpha)^{13}N$	10 min
$^{16}O(p,3p)^{14}C$	5770 years
$^{16}O(d,\alpha)^{14}N$	Stable†
$^{16}O(p,pn)^{15}O$	124 sec
$^{16}O(p,2p2n)^{13}N$	10 min
$^{16}O(p,3p3n)^{11}C$	20.5 min
$^{16}O(^{3}He,\alpha)^{15}O$	124 sec
$^{16}O(d,t)^{15}O$	124 sec
$^{16}O(\alpha,p)^{19}F$	Stable*

*Proton is measured.
†Alpha is measured.
‡0.875-Mev prompt gamma is measured.

Ricci et al.[23] reported an improvement in the ^{3}He activation analysis procedure introduced by Markowitz and Mahony.[24] In Ricci's work a Van de Graaff accelerator was used to produce ^{3}He ions with energies of up to 10 Mev. Analyses were carried out for surface oxygen on samples of Fe, Ge, Si, and Be. An ultimate sensitivity of 2×10^{-10} g oxygen was reported by measurement of the ^{15}O from the $^{16}O(^{3}He,\alpha)^{15}O$ reaction and the ^{17}F nuclide from the $^{16}O(^{3}He,pn)^{17}F$ reaction. For ^{18}F a sensitivity of 3×10^{-11} g was obtained. Ricci listed sensitivities and detection limits both for thick targets that contained oxygen and for thin foils that contained low-Z elements; the lowest limit shown in the first case was 2.35 ppb by the $^{16}O(^{3}He,p)^{18}F$ reaction, and in the second, 28.6 ppb by the same reaction.

Demildt[25] has found the ^3He activation analysis method ideal for determining oxygen in actinide metals. By extrapolating from available data, he estimated that oxygen can be determined in as low a concentration as 0.001% in a few hundred micrograms of matrix material. Interference by activation of other elements can be restricted by control of the bombarding energy of the ^3He particles. This restriction makes the method applicable to the analysis of fissionable actinides since no nuclear reactions will occur between the target matrix and the bombarding particles if the kinetic energy is lower than the coulomb barrier.

DETERMINATION OF O_2 IN SOLUTIONS AND GASES

The determination of oxygen in solutions and gases is not considered as severe a problem as the determination of oxygen in oxides. The difficulty is eased because oxygen is present either in the dissolved form or as a mixture and is not chemically bound. With solutions, and particularly in seawater, the classical Winkler method is still used which is based on the oxidation by oxygen of Mn(II) to Mn(IV) which, in turn, can oxidize iodide to iodine in acid solution. The resulting iodine can then be titrated with thiosulfate to give an indirect method for monitoring the oxygen content.[26] The equations for the reaction are:

$$Mn(OH)_2 + O \rightarrow MnO(OH)_2$$

$$MnO(OH)_2 + 4H^+ + 3I^- \rightarrow Mn^{2+} + I_3^- + 3H_2O$$

$$I_3^- + 2S_2O_3^{2-} \rightarrow 3I^- + S_4O_6^{2-}$$

More recently, various electroanalytical methods have come into use.[26] In voltametric methods the oxygen undergoes a reaction at an electrode surface held at a fixed potential. The current flow that results from the electrode reaction can be directly related to the quantity of oxygen present in the test solution. Other methods—colorimetric and titrimetric—rely upon the relatively high oxidation potential of oxygen to produce a colored product or a substance that can be measured by a redox titration.

With gases the paramagnetic properties of the oxygen molecule may be used for its determination. Mass spectrometry can be used for the determination of oxygen in a gas stream and to differentiate between the isotopic species that may be present. The technique of gas chromatography may be used to separate and determine quantitatively the oxygen from a gas stream. The electrometric methods that are used for determining gas in solutions may be used for determining oxygen in gas streams provided that the gas stream is allowed to bubble through a solution or to impinge upon a semipermeable membrane and to penetrate into a moistened electrolyte.

Currently, considerable attention has been focused on oceans and lakes not only from the purely scientific aspect but also from the standpoint of increasing the available food supply of the world. One of the important parameters to be taken into account is the oxygen content of the water at various locales and depths. A proposed method for determining the oxygen content of fresh- and seawater is based on the quantitative reaction between oxygen and a radioactive material. The amount of radioactivity released in the test water sample or effluent of a flow-through-type cell is a measure of the oxygen content of the water. Procedures such as that described above have become known as "radio-release" methods.[27]

Richter and Gillespie[28] suggested such a scheme in which the oxygen contained in the water oxidizes metallic thallium to the soluble Tl(I) ion,

$$4Tl_{(s)} + O_{2(aq)} + 2H_2O_{(liq)} \rightarrow 4Tl^+_{(aq)} + 4OH^-_{(aq)}$$

Thallium labeled with ^{204}Tl was plated on copper turnings for use in freshwater, or on molybdenum for seawater. The solid metal was then placed in contact with the water sample that contained oxygen. After a time interval sufficient to allow completion of the reaction, the sample was removed and the activity determined by counting. The oxygen content was directly related to the counts obtained from the "released" activity in the solution. In seawater a nonlinear response was obtained above 3 ppm, and the use of a calibration curve was necessary. Further development of a complete instrument was reported by Gillespie and Roberts.[29]

With ^{204}Tl of specific activity 2 mc/g, the limit of detection was 0.2 ppm O_2; the use of ^{204}Tl as obtained from Oak Ridge National Laboratory would increase the sensitivity to a limit of detection of ~0.1 ppb.

Ross,[30] in a recent investigation of the oxygen content of seawater, modified the Winkler method to allow a radio-release final step. He tagged the iodide that was added to the test solution with radioactive iodine. The iodine ultimately representing the oxygen content of the seawater was quantitatively extracted with chloroform. The chloroform solution was then counted to determine the iodine content. As little as 0.5 μg oxygen can be determined according to Ross.

Tracer Techniques

THE DETERMINATION OF TRACER OXYGEN

Oxygen takes part in almost all biological processes by way of cellular respiration or through the formation of water by hydrolytic processes. The use of tracer isotopes offers the possibility of following the course of such reactions, but the radioactive isotopes of oxygen have such short half-lives that the possibility is negated except for a few cases. The stable isotopes ^{18}O and ^{17}O may be used for labeling compounds for the purpose of following metabolic processes. These stable isotopes in the past have been identifiable only through mass spectrometry. Fleckenstein and Janke[31]

introduced the use of the irradiation of ^{18}O with 4.05-Mev protons to produce radioactive ^{18}F. They illustrated the method by following the course of organic phosphoryl compounds in intracellular processes. The compounds were separated by paper chromatography, and the portion of the paper containing the isolated compound was homogeneously dispersed and spread in a thin layer on platinum foil. The foil was exposed to a proton beam of 4.05 Mev energy in a cyclotron and the ^{18}F activity counted as a measure of the ^{18}O present. Fleckenstein and Janke recommend the activation procedure in preference to the use of mass spectrometry.

Ellis[32] reviewed previous work in this field and suggested a neutron activation determination of ^{18}O to be followed by a chemical separation. Ellis' method involved the release of the oxygen as carbon dioxide and subsequent conversion into ammonium carbamate prior to neutron activation. Irradiation of ^{18}O with thermal neutrons yielded ^{19}O with a half-life of 29.5 sec. The ^{19}O decayed by beta emission to ^{19}F, which emits gamma rays in the transition to the ground state. The gamma-ray activity was measured by scintillation spectrophotometry. Ellis also tabulated other nuclear reactions that may be used for the activation analysis of ^{18}O. He concluded that with further refinement the carbamate conversion method holds promise of becoming competitive with mass-spectrometric techniques for ^{18}O determination.

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A Compilation of Beta-Excited X-Ray Spectra*

By F. J. Miller

Abstract: A selected number of beta-excited X-ray sources were fabricated and their spectrums compiled. Thirty-one target elements were used, with ^{147}Pm, ^{45}Ca, and ^{32}P as the beta emitters. Standardization was maintained by holding the assay parameters, such as the detectors and the geometry, constant. The parameters were recorded throughout so that spectrums from variants of a single source type could be compared to each other, as could different source types. The basic source types used were the apposition, the mixture, and the compound. The compilation is intended to provide a convenient and rapid qualitative as well as quantitative reference for the worker using the beta-excitation phenomenon.

In the simplest terms, beta-excited X-ray sources are made of a small amount of beta-emitting radioisotope and a thin covering layer of target material. The action of the radiation on the target material results in an emission spectrum that includes the bremsstrahlung photon continuum due to beta-ray acceleration and the superimposed characteristic X-ray emission of the target atom (most usually the K_α X ray).

Bizzell and Hitch[1] first demonstrated in 1951 that the beta-excited photon source had practical application by their measurement of silt density in the sediment of TVA lakes. More recently the technique has been used for analytical absorptiometry and X-ray fluorescence. The apparatus is compact, light, and extremely stable, and the source requires no power input. The small radiative power output of the source is no handicap to analytical uses such as determining the content of lead nitrate in water, lead tetraethyl in gasoline, or alcohol in water. The polychromatic nature of the photon beam is a disadvantage for absorptiometry, but satisfactory results have been obtained in the past with similar beams. Proper choice of emitter and target can provide beams that are nearly monochromatic. The potential usefulness of beta-excited photon sources has engendered interest in their design and manufacture.

A knowledge of the emission spectrums obtainable from beta-excited photon sources is needed to design a source best suited for a specific purpose. The spectrums available in the literature are not coherent because individual workers have used various geometries, different source types, and any number of types of photon detectors and modes of operation. The intercomparison of these spectrums is difficult at best. The need for a compilation of sets of spectrums produced under controlled and standardized conditions by the more common beta-excited X-ray sources has been filled by this work, supported by the USAEC Division of Isotopes Development.

The project was planned to compile a set of standard spectrums produced by the important beta emitters fabricated in the primary geometries usually used for beta-excited X-ray sources. Some 22 target elements were selected and studied in combination with 15 pure-beta emitters. The types were limited to the apposition source (in which the beta emitter is sandwiched between two layers of target material), the mixture source (in which the beta emitter is intimately mixed with the elemental target in powdered form), and the compound source (in which the beta emitter is combined with the target element into a molecule or mixed with a compound of the target element).

The compilation is being issued in three parts. Part I, covering ^{147}Pm, ^{45}Ca, and ^{32}P, has been published; Part II, which will cover ^{63}Ni, $^{90}Sr-^{90}Y$, and ^{204}Tl, is in press; and Part III, covering ^{143}Pr, ^{89}Sr, ^{14}C, ^{3}H, ^{35}S, and ^{99}Tc and containing a bibliography, will probably be issued in 1967.

Beta-Excitation Mechanism

Beta-excitation phenomena occur whenever beta particles impinge on a target atom. Ideally, for use in a beta-excited X-ray source, the beta-emitting radionuclide should emit no other radiation. The

*L. E. Preuss, H. Collins, C. Artman, and C. Bugenis, A Compilation of Beta Excited X-Ray Spectra, Part I and Foreword (^{147}Pm, ^{45}Ca, ^{32}P), USAEC Report TID-22361 (Pt. 1), Henry Ford Hospital, Detroit, issued November 1966 (available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Virginia 22151, $3.00, 202 pp.).

target material may be a highly purified target element, several target elements may be used, or the radioactive nuclide may be chemically combined with the target element.

Ionization and bremsstrahlung production, two processes which are of primary interest for beta-excited X-ray sources, occur when beta rays are incident upon matter. Ionization takes place when an electron, as a result of collision with an incoming beta particle, is ejected from an inner electron shell. The ejected electron is replaced by an electron from an outer electron shell, and the difference in energy between the orbital electron shells appears as a quantum of energy that is released as an X-ray photon. The energy of the photon is characteristic of the particular orbital electron shells for each specific atom, and the X-ray photon is denoted by the element and the lower energy electron shell involved. Bremsstrahlung is produced when the electrical field of the target nucleus is disturbed by the near passage of a beta particle and, as a result of the interaction, the velocity of the beta particle is changed. The acceleration of the beta particle causes the emission of radiation that is continuous over a spectral range and does not exhibit the sharp energy peaks of characteristic X rays. The radiation spectrum emitted by the target atom is a composite of the continuous, "white," radiation of bremsstrahlung and the superimposed peak or peaks of the characteristic X rays.

The radionuclide itself does not remain unaffected by the emission of the beta particle, although the radiations produced make up only a small part of the total spectrum of the beta-excited X-ray source. X rays and Auger electrons are produced as a result of internal ionization of the emitting atom. Internal bremsstrahlung, also a continuous radiation, is produced in the beta-emitting atom as a result of the sudden change of its electric charge during beta emission.

When monoenergetic electrons are stopped by "thick" absorbers, the bremsstrahlung energy output, I, may be described by

$$I = kZE^2 \qquad (1)$$

where I = total bremsstrahlung energy, Mev
$E = 0.5(T/m_0c^2)$, Mev, where T is the kinetic energy of the impinging particle, m_0 the electron mass, and c the velocity of light
k = a constant in units of Mev^{-1}
Z = atomic number of the target element

From this expression, similar relations may be developed to predict the average external bremsstrahlung per average beta.

The shape of the spectral distribution of bremsstrahlung can be expressed by

$$\frac{dI}{h\nu} = \frac{1.02\ kZ \int_{W_1}^{W_0} (W - W_1)\ N(W)\ dW}{\int_{W_1}^{W_0} N(W)\ dW}$$

where W_0 = maximum beta energy, E_0, in m_0c^2 units
$W_1 = h\nu/0.51 + 1$
$W = E/0.51 + 1$

Attenuation of the low-energy end of the spectrum results in a deviation of the shape from that predicted.

Beta-Excited Source

Table 1 lists 12 pure-beta emitters that may logically be used to excite X rays on the basis of availability, half-life, beta energy, existence of radioactive daughter, and lack of radioactive contaminants.

Cataloging the spectrums formed by combination of all the elements with the listed pure-beta emitters is impracticable; therefore several target elements were selected to provide a reasonable range of atomic numbers, as shown in Table 2. Of the 33 listed target elements, 22 can be obtained in foil, powder, and compound forms. In the compilation, each pure-beta emitter in combination with 33 elements is presented in a separate section. Each complete section lists 22 beta-source/target combinations and shows the spectrums for apposition, mixture, and compound targets. Additionally, 11 spectrums are presented for mixture sources alone.

PREPARATION OF BETA SOURCES

The three types of sources are made as follows:

Apposition Source: A solution of a beta emitter is pipeted onto a backing-target foil. After the deposited material has been dried, a thin covering foil of the same material as the backing is sealed to the backing with epoxy glue. The thickness of the backing foil is held constant at 5 mils, but that of the covering foil ranges from 0.2 to 0.5 mil. A low-atomic-number absorber, known as the "beta stop," is placed on top of the source to absorb all beta radiation.

Mixture Source: The powdered mixture is placed in a Lucite vial, the bottom of which has been ground to a thickness just sufficient to stop beta radiation.

Compound Source: The beta source and the target material are chemically combined and placed in a Lucite vial, the bottom of which is ground as that of the mixture source.

The pure-beta emitters used in the study were calibrated against (1) an NBS standard and (2) a commercial standard; when these were not available, the calibration value supplied by Oak Ridge National Laboratory (ORNL) was accepted, or zero-absorption calibration was carried out. The last method was used, when possible, in preference to acceptance of the ORNL value; in the zero-absorption method, a standard is prepared, consisting of a pure-beta emitter of known activity deposited on a copper planchet. It is counted by a Geiger tube with a series of aluminum absorbers, and the absorption curve is extrapolated to zero. The test beta emitter is pre-

Table 1 PURE-BETA EMITTERS

Isotope	$E_{max.}$, Mev	Half-life	Production	Activity,* curies/g	Purity of available radionuclide, %	Daughter†
$^{14}_{6}C$	0.155	5568 years	$^{14}N(n,p)^{14}C$	69.0 at.% ^{14}C‡	> 95	^{14}N
$^{45}_{20}Ca$	0.254	164 days	$^{44}Ca(n,\gamma)^{45}Ca$	0.500	> 99	^{45}Sc
$^{45}_{20}Ca$	0.254	164 days	$^{44}Ca(n,\gamma)^{45}Ca$	17.000	> 99	^{45}Sc
$^{3}_{1}H$	0.018	12.262 years	$^{6}Li(n,\alpha)^{3}H$	1.030§	–	^{3}He
$^{63}_{28}Ni$	0.067	125 years¶	$^{62}Ni(n,\gamma)^{63}Ni$	12.119	> 95	^{63}Cu
$^{63}_{28}Ni$	0.067	125 years¶	$^{62}Ni(n,\gamma)^{63}Ni$	1.425‡	–	^{63}Cu
$^{32}_{15}P$	1.707	14.22 days	$^{32}S(n,p)^{32}P$	C. F.**	> 99 (Excl. of ^{33}P)	^{32}S
$^{147}_{61}Pm$	0.223	2.64 years	Fission	C. F.	> 99 (Traces of ^{241}Am)	^{147}Sm
$^{147}_{59}Pr$	0.932	13.76 days	Fission	C. F.	> 98	^{143}Nd
$^{35}_{16}S$	0.167	87 days	$^{35}Cl(n,p)^{35}S$	C. F.	> 99 $^{32}P < 0.1$	^{35}Cl
$^{89}_{38}Sr$	1.463	50.5 days	Fission	C. F.	> 99 ($^{90}Sr - ^{90}Y < 10$)	^{89}Y
$^{90}_{38}Sr - ^{90}_{39}Y$	0.545 (Sr) 2.260 (Y)	27.7 years 64.2 hr	Fission Fission	C. F. C. F.	> 99 ($^{89}Sr < 10$)	^{90}Zr
$^{99}_{43}Tc$	0.290	2.12×10^5 years	Fission	0.020	> 99	^{99}Ru
$^{204}_{81}Tl$	0.764	3.56 years	$^{203}Tl(n,\gamma)^{204}Tl$	1.000	> 99	^{204}Pb (98%), ^{204}Hg (2%)

*Obtained from Oak Ridge National Laboratory, Oak Ridge, Tenn.

†For all but ^{147}Pm, decay is to the ground level of the stable daughter. The ^{147}Pm daughter, ^{147}Sm has a half-life of 1.3×10^{11} years and makes only a negligible contribution.

‡Atomic Energy of Canada Ltd., Ottawa.

§Volk Radiochemical Co., Skokie, Ill. (now, Division of Nuclear Consultants Corp.).

¶Corrected value 92 years; cf., G. Goldstein and S. A. Reynolds, Specific Activities and Half-lives of Common Radionuclides, *Nuclear Data*, A, 1: 435-452 (1966).

**Carrier free.

Table 2 TARGET ELEMENTS — LISTED ACCORDING TO SOURCE GEOMETRY

Apposition	Mixture	Compound
Mg	Mg	Mg
Al	Al	Al
Ti	Ti	Ti
V	V	V
Fe	Fe	Fe
Co	Co	Co
Ni	Ni	Ni
Cu	Cu	Cu
Zn	Zn	Zn
Zr	Zr	Zr
Nb	Nb	Nb
Mo	Mo	Mo
Pd	Pd	Pd
Ag	Ag	Ag
Cd	Cd	Cd
In	In	In
Sn	Sn	Sn
Ta	Ta	Ta
W	W	W
Pt	Pt	Pt
Au	Au	Au
Pb	Pb	Pb
	Si	
	Cr	
	Ge	
	As	
	Se	
	Ru	
	Sb	
	Te	
	Os	
	Ir	
	Bi	

pared and counted in exactly the same way, and its absorption curve is also extrapolated to zero. The test activity is then equal to the ratio of the zero absorption values multiplied by the activity of the standard.

Activities of 200 μc or less were used for convenience in preparing the source and obtaining the compiled spectrums. For a practical source the amount of activity would have to be scaled up by a factor of 10^4. Caution should be observed in extrapolating the compiled values to the amount of activity required for a practical source since artifacts may be introduced. Self-absorption within the larger amount of active material and the possible presence of contaminating heavy metals will cause a deviation from the extrapolated value.

The apposition source is of more interest than the other two types of sources since the intensity of the peaks in the emitted spectrum may be changed by the addition or removal of transmission target layers. Additional target layers may be provided either by vacuum deposition of metals or by use of thin foils. In a study of the effect of target material thickness on K_α photon intensity, step thicknesses of 0.01-mil silver on a promethium beta emitter were used. An initial rapid rise in K_α photon production was obtained until a plateau was reached at 1.4×10^5 A, after which a decrease in intensity was noted.

The substantial photon contribution made by the backing target, whether of the same or different ma-

terial from the transmission target, should be clearly realized by possible users of the compiled data for the design of practical sources, especially since the backing support of an apposition source exhibits an effect not seen in either of the other types of sources. If a backing target of an element different from that of the covering target layer is used, the backing target will add its K_α X-ray component as well as its white continuum to the overall spectrum. If the backing support is of the same material as the transmission target layer, the photon emission of the transmission layer will simply be enhanced by the backing contribution. The backing target thickness is a critical factor in determining its photon contribution. Thickness beyond the range of the beta particle is wasted; the maximum photon output is obtained by making the backing thickness approximately one-third the beta range.

PHOTON DISPERSION FROM BETA-EXCITED SOURCES

Anisotropic photon directivity from the practical beta-excited source has to be taken into account in the use of these photons. Each type of source has a specific geometry, which influences its photon dispersion pattern. As a further complication, the dispersion of the characteristic X rays differs from that of the bremsstrahlung. An even more critical factor to be considered is the absorption mechanism in the source and in the beta stop.

Source directivity is especially evident in the apposition source because of its disklike geometry. Figures are presented in the compilation that display in polar coordinates the K_α and bremsstrahlung dispersion from different apposition sources.

The right-cylinder geometry of the mixture and compound sources creates an X-ray dispersion pattern for these that differs from that of the apposition source. Plots are given in the compilation that show the dispersion pattern for all photons (K_α X rays and bremsstrahlung). In the design of practical sources, factors contributing to the dispersion patterns of the different types of sources must be considered.

Instrumentation

STANDARDIZATION AND REPRODUCIBILITY

Methods, detectors, analysis instrumentation, and associated electronic systems were chosen for simplicity, uniformity, and availability so that users of the compilation could easily reproduce the compiled spectral measurements. A block diagram of the system is shown in Fig. 1.

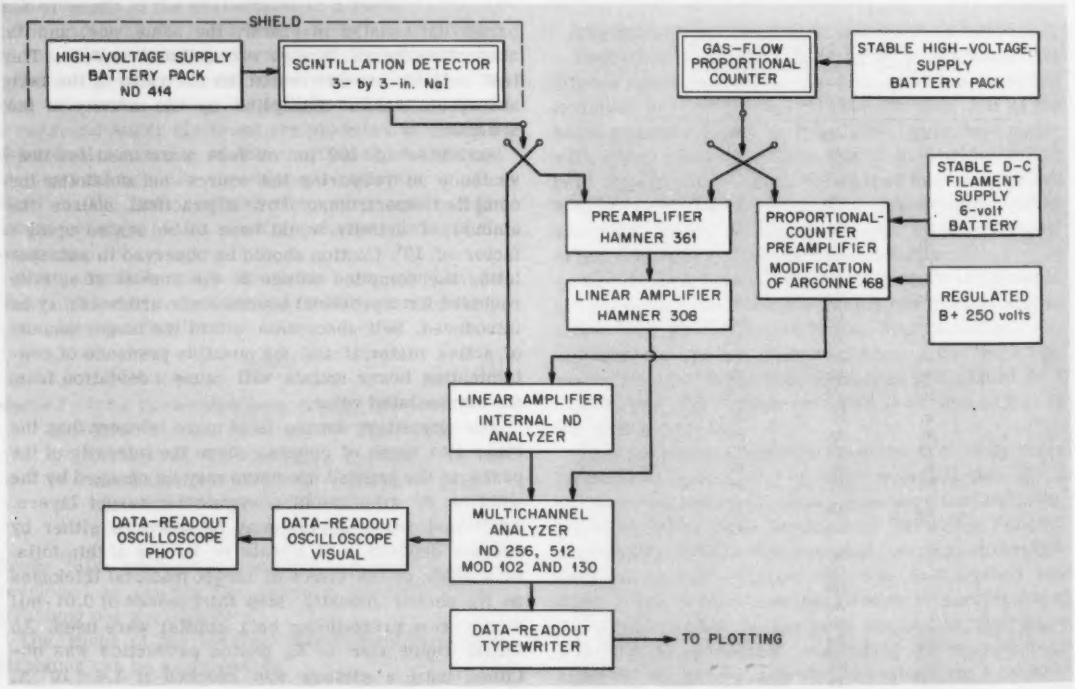
SHIELD
HIGH-VOLTAGE SUPPLY BATTERY PACK ND 414
SCINTILLATION DETECTOR 3- by 3-in. NaI
GAS-FLOW PROPORTIONAL COUNTER
STABLE HIGH-VOLTAGE-SUPPLY BATTERY PACK
STABLE D-C FILAMENT SUPPLY 6-volt BATTERY
PREAMPLIFIER HAMNER 361
PROPORTIONAL-COUNTER PREAMPLIFIER MODIFICATION OF ARGONNE 168
REGULATED B+ 250 volts
LINEAR AMPLIFIER HAMNER 308
LINEAR AMPLIFIER INTERNAL ND ANALYZER
DATA-READOUT OSCILLOSCOPE PHOTO
DATA-READOUT OSCILLOSCOPE VISUAL
MULTICHANNEL ANALYZER ND 256, 512 MOD 102 AND 130
DATA-READOUT TYPEWRITER
TO PLOTTING

Fig. 1 Layout of the amplification and analysis network used for collection of the data in the compilation. The integral preamplifiers and analyzer internal linear amplifier were used primarily to collect data. The Hamner 361 and 308 amplifiers were used for check of base-line shift.

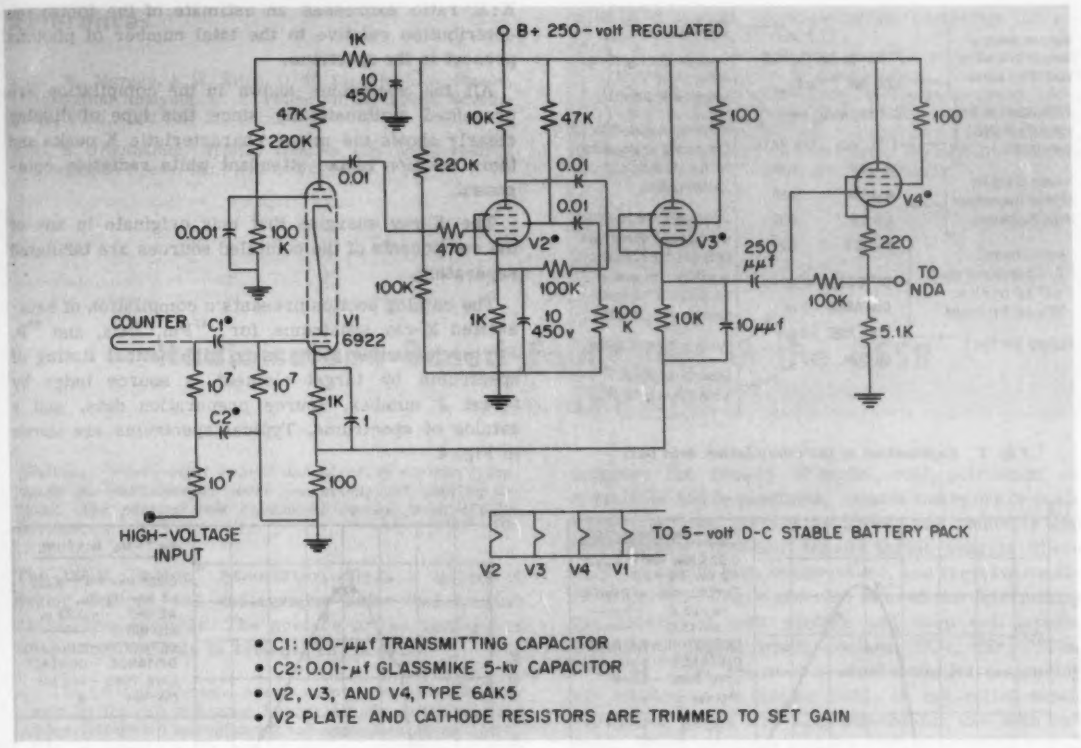

Fig. 2 Adaptation of the Argonne model 168 preamplifier used for the flow-gas counter in the spectrum tests.

A crystal and a proportional-counter detector were used to collect spectral data. Both of these were supplied with high voltage from a battery pack. The detectors shared a common pulse-amplification and analysis network that consisted of either a Hamner 361 and 308 linear amplifier or the integral linear amplifier system of the ND-130 analyzer.

The pulse analysis system was made of commercially available components. The linear preamplifier used with the gas-flow proportional counter was an Argonne model 168 adapted, as shown in Fig. 2, to the specific use required.

Either a NaI(Tl) scintillation detector or a gas-flow proportional counter was used for photon detection. The scintillation detector with a 3- by 3-in. NaI(Tl) crystal, used with the phototube operated at 700 kev, had a resolution of 7.3% for the ^{137}Cs peak. The 3- by 3-in. crystal was used for counting sources when the energy of the characteristic radiation was 17 kev or above; a smaller crystal was used for the 10- to 17-kev region.

The gas-flow proportional counter used was similar to that described by Crouthamel.[2] The preamplifier and anode high-voltage input were located at opposite ends of the counter in shielded chassis boxes. The counter and preamplifier combination was used in the spectral region down to 2 kev although it operates best in the 4- to 20-kev region. Reference should be made to basic texts[2-4] for equipment calibration information.

COMPILATION DATA AND INTERPRETATION

In the compilation, spectrums collected by the detector and analyzer systems are presented. No correction other than subtraction of background was made. Necessary information is presented in case the user wishes to make other corrections.

The data are presented so that optimum clarity in reading the spectrums is obtained. Each curve is drawn by the draftsman to provide the best fit, and each fifth channel point is plotted exactly as it occurs. For areas under each of the K peaks, each channel point is plotted.

The ratio of the area under the K_α X-ray peak to the total area under the entire spectrum is denoted as $K:\Sigma$, and $K:\Sigma$ data are provided in the corner of each spectrum plot. A typical data box with explanation of other compiled data is shown in Fig. 3. The

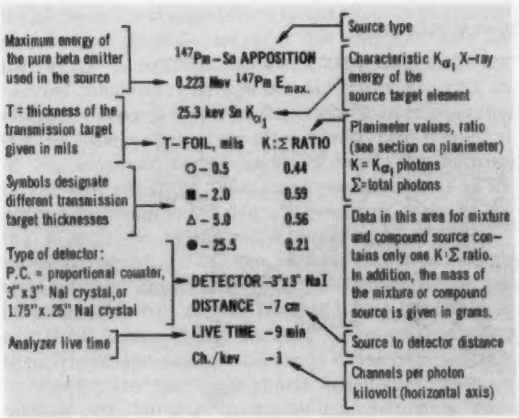
Maximum energy of the pure beta emitter used in the source
147Pm–Sn APPOSITION
0.223 Mev 147Pm Emax.
Source type
Characteristic Kα1 X-ray energy of the source target element
25.3 kev Sn Kα1
T = thickness of the transmission target given in mils
T–FOIL, mils K:Σ RATIO
Planimeter values, ratio (see section on planimeter)
K = Kα1 photons
Σ = total photons
Symbols designate different transmission target thicknesses
O – 0.5 0.44
□ – 2.0 0.59
Δ – 5.0 0.56
● – 25.5 0.21
Data in this area for mixture and compound source contains only one K:Σ ratio. In addition, the mass of the mixture or compound source is given in grams.
Type of detector: P.C. = proportional counter, 3"x3" NaI crystal, or 1.75"x.25" NaI crystal
DETECTOR – 3"x3" NaI
DISTANCE – 7 cm
Source to detector distance
Analyzer live time
LIVE TIME – 9 min
Ch./kev – 1
Channels per photon kilovolt (horizontal axis)

Fig. 3 Explanation of the compilation data box.

K:Σ ratio expresses an estimate of the ionization contribution relative to the total number of photons present in the spectrum.

All the spectrums shown in the compilation are presented arithmetically since this type of display clearly shows the minor characteristic K peaks and their relation to the attendant white radiation component.

The X-ray energies that may originate in any of the components of the compiled sources are tabulated separately.

The catalog section presents a compilation of beta-excited X-ray spectrums for ^{147}Pm, ^{45}Ca, and ^{32}P. For each nuclide there is an alphabetical listing of spectrums by target element, a source index by target Z number, source preparation data, and a catalog of spectrums. Typical spectrums are shown in Fig. 4.

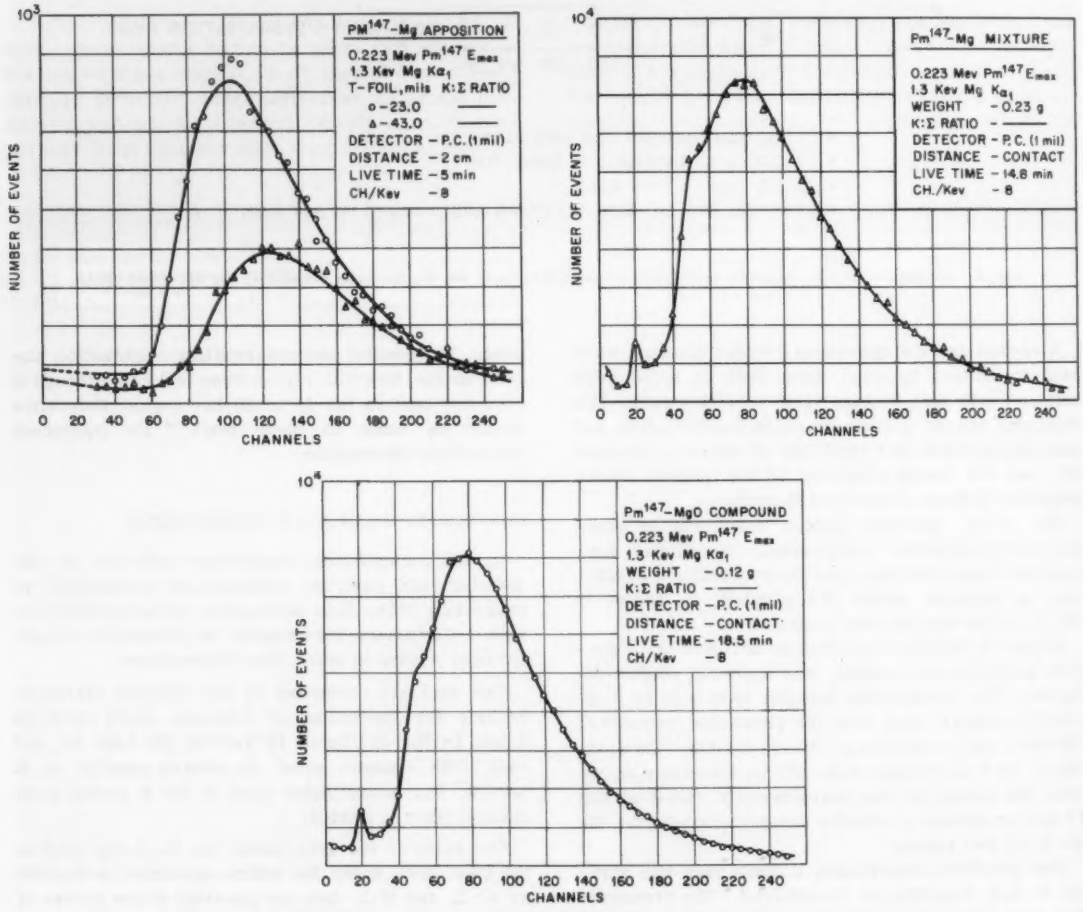

Fig. 4 Examples of spectrums produced by apposition, mixture, and compound sources.

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Radioisotope-Based Equipment in the USSR

By Martha Gerrard

Abstract: Forty-eight relays and gages, of various types, based on radioisotopes were marketed last year in the USSR. The cost of this equipment varies from $250 to $11,000.

The USSR "Izotop" Association offers a variety of gaging devices and radiography units that contain radioactive nuclides. The preface to the listing[1a] of equipment available in 1966 states, in part:

> The All-Union Isotope Association of the State Commission for the Use of Atomic Energy is made up of organizations interested in equipment and apparatus whose operation is based on the use of nuclear radiation sources. Radioisotope equipment is safe to use and in many cases cannot be replaced by equipment that operates on other principles. It is universal and can be used in various systems for automatic monitoring and control of industrial processes without contact of the unit with the medium being monitored. The association is also concerned with gamma-radiography apparatus for detecting leaks in welded and cast metal parts under field and factory conditions.

Izotop installs, adjusts, repairs, and services the equipment it makes. In addition, it installs experimental and pilot models of radioisotope equipment upon request of a manufacturing plant and offers consulting services.

Earlier equipment catalogs[1b,c] describe many of the same units that were listed in the 1966 newsletter, which indicates that these have stood up in use. Of course, some of those listed earlier are no longer carried. The association also issues pamphlets that describe in detail[2] the equipment and its use. Many of the USSR radioisotopic instruments are described in a recently translated book.[3] In a 1964 review[4] of some of the USSR radioisotope equipment, the annual saving resulting from its use in five different industries was stated to be 240,000 rubles ($264,000).

The 1966 catalog lists 48 pieces of radioisotope equipment. Level gages containing ^{137}Cs or ^{60}Co can remotely measure the level of oil and oil products in reservoirs and control the charge level in a blast furnace. Density gages, which also use ^{137}Cs or ^{60}Co, measure the density of metal, coal, petroleum, or chemicals inside pipelines; remove heavy ore or coal-lump fractions from classifiers; and remotely and continuously measure density inside vessels where high-pressure, high-temperature, and abrasive conditions prevail. Gages are also offered for determining the density of both surface and deep soil layers. Thickness gages, which contain ^{144}Ce, ^{90}Sr, ^{170}Tm, ^{204}Tl, or ^{147}Pm, are used in steel mills for measuring the thickness of moving cold- or hot-rolled steel. Thickness gages are also offered for use with organic films or paper and for determining the thickness of coatings whose atomic number differs from that of the base material, e.g., paint, galvanized coatings, or even of tin on either side of a steel ribbon. Irregularities in fibers, yarn, or fabrics in textile mills may be controlled with a device that contains four ^{204}Tl sources.

Plutonium or polonium with beryllium form the neutron sources for gages that measure moisture in surface or subsurface soil.

Radiography units are made with ^{137}C, ^{192}Ir, ^{170}Tm, or ^{60}Co. A so-called "universal" radiography unit uses one of these, or ^{75}Se, or ^{152}Eu.

Equipment for analytical purposes includes apparatus containing a ^{55}Fe source that can determine the sulfur content of diesel fuel. A two-component liquid analyzer, for use with aqueous methyl alcohol solutions, determines the percentage of alcohol in the solution by measuring the absorption of a ^{90}Sr beam. A three-component solution analyzer, which also uses ^{90}Sr, measures the sodium hydroxide (80 to 100 g/liter) and sodium chloride (160 to 220 g/liter) concentrations in aqueous electrolytic solutions.

Some of the other apparatus offered are an object counter, a temperature regulator, a device for continuous weighing of nonmetallic minerals on a conveyor belt, and a smoke detector as well as some auxiliary equipment. An interesting piece of equipment for measuring the thickness of a layer of coal dust was offered in 1965 but withdrawn in 1966.

The cost of this equipment varies from $250 for a gamma relay for level measurements to $11,000 for the most elaborate level-following gage. Some other costs are $2200 for a read-through thickness gage and $520 for a beta-backscatter coating-thickness gage. Industrial radiography units run from $700 to $2000, depending on the source strength. The diesel fuel analyzer costs $2800, and the three-component-liquid analyzer, $4400.

Equipment Under Development

Each of Izotop's catalogs describes or lists equipment under development. The listing[1a] for 1966 mentioned an explosion-proof level indicator (^{60}Co, $550). It is designed to regulate levels of filling of bins and reservoirs with liquids or other free-flowing materials in the mining, coal, electrical, metallurgical, oil, chemical, and other industries. A portable device for detecting methane in mines contains tritium ($440). Several new thickness gages have been designed: (1) for hot-rolled sheet (^{137}Cs, $16,500); (2) for rolled metal of 800 g/m^2 (^{147}Pm for up to 150 g/m^2, ^{204}Tl for 0 to 800 g/m^2) and of 9000 g/m^2 (^{90}Sr, ^{144}Ce, or ^{106}Ru, depending on the range); and (3) for rolled steel, copper, nickel, zinc, and their alloys up to 3 mm and titanium up to 5 mm [^{170}Tm or ^{90}Sr (bremsstrahlung)].

Safety

Izotop requires completion of detailed questionnaires on the specifications, conditions of use, and design of the process to be controlled before production of a device is started. The completed questionnaire constitutes, in effect, a sales contract. In accordance with USSR regulations on the use of radioactive materials, the acquisition of radioisotopic control and measurement devices does not require prior approval of local public health officials. However, the devices are delivered with instructions for safe installation and operation, and local health officials must be satisfied that the instructions have been followed before operation is permitted. Conditions that must be fulfilled include limiting the radiation from the source unit to a maximum of 10 mr/hr at the surface and 0.3 mr/hr at 1 m. For portable gamma-radiography units the radiation at 1 m from the housing surface must not exceed 3 mr/hr, although for stationary units the allowance is somewhat higher. In any case the radiation dose to service personnel must not exceed 0.1 r/week.

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Puerto Rico Nuclear Center: Its Principal Irradiation Facilities and Scientific Program

By F. E. Rushford*

Abstract: The Puerto Rico Nuclear Center is well equippea with irradiation facilities to conduct a diversified program of research and training in the peaceful applications of nuclear energy. Research is directed chiefly toward problems of national and international interest which are best approached in the tropical environment such as that of Puerto Rico.

The Puerto Rico Nuclear Center (PRNC), established in October 1957, is operated for the U. S. Atomic Energy Commission by the University of Puerto Rico (UPR). The objective of the PRNC is to offer comprehensive programs for advanced training and research in the peaceful applications of nuclear energy in agriculture, medicine, engineering, and science. In support of formal education, the PRNC assists in the graduate and professional programs of the University and offers to qualified students opportunities for research at the master's, doctorate, and postdoctorate levels. Part of the basic mission of the PRNC is to offer opportunities for Latin Americans to study the peaceful applications of nuclear energy and to promote the utilization of this technology in the development of their respective countries. English and Spanish are spoken by the majority of the staff.

Irradiation Facilities

The chief irradiation facility at the PRNC is an AMF pool-type research reactor (Fig. 1), currently operated 16 hr/day at a 1-Mw power level. It is a light-water-moderated heterogeneous solid-fuel reactor cooled with water. The water-filled concrete pool is in two sections, and the reactor core may be placed in either section. One section contains an experimental stall in which beam tubes and other experimental facilities converge, and the other is an open area permitting bulk irradiation. Two 8-in.-diameter and four 6-in.-diameter beam tubes extend in horizontal planes outward from the reactor core. Samples are carried into the high-neutron-flux areas at the core by a pneumatic system operated by a constant-exhaust system of concentric aluminum air lines.

A thermal column—a stacked graphite and lead assembly—is provided for experiments with highly thermalized neutrons. The pneumatic tube flux is 10^{12} n/(cm^2)(sec); that of the thermal column varies but is approximately 10^8 n/(cm^2)(sec).

An integral part of the pool construction is a gamma-irradiation room where highly radioactive fuel elements may be used as a radiation source. The room is square at the top, 6 ft on an edge, and the walls taper to an aluminum window, 2 ft square, at a depth of 27 ft. This window is the only partition between the room and the reactor pool.

An L-77 homogeneous aqueous uranyl sulfate reactor with a beam-tube flux of 10^8 n/(cm^2)(sec) is used principally for training in the graduate nuclear engineering program.

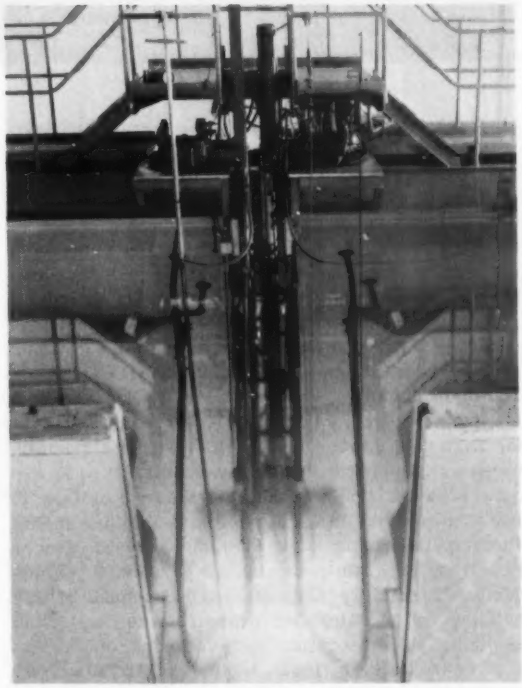

Fig. 1 A PRNC pool-type reactor core suspended from a manually operated bridge.

A natural uranium–graphite subcritical assembly is also available.

The main ^{60}Co gamma-irradiation facility (Fig. 2) is housed in a 9- by 8- by 14-ft-deep water pool. A 3- by 8-ft platform located 10 ft below the water surface holds the variable-geometry irradiator. On the platform are 12 tubes that hold the 8-in.-diameter ^{60}Co capsules containing approximately 200 curies each. The radiation field is accurately reproducible. The

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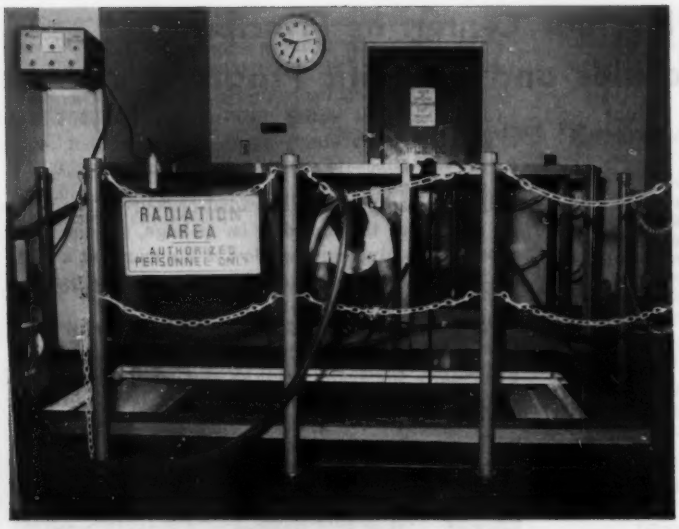


Fig. 2 A PRNC ^{60}Co gamma-irradiation facility.

10-ft depth is sufficient to decrease the radiation level at the surface to less than 0.2 mr/hr but not too deep for ready observation of operations. Most biological samples are irradiated in wide-mouthed polyethylene bottles that are weighted to overcome buoyancy. To the screw tops of the bottles are attached aluminum rods to facilitate underwater handling. The tops also have two hose connections—for air that is maintained at a positive pressure inside the bottle to exclude water, or for oxygen or nitrogen to allow irradiations in different atmospheres. A sealed metal Dewar flask, 15 cm in diameter, containing ice has been used for low-temperature irradiations. A thermocouple inside the flask with a recording gage above the pool permits monitoring temperatures during irradiation.

An 8000-curie ^{60}Co medical teletherapy unit (Fig. 3) installed in the PRNC Bio-Medical Building in Río Piedras is used in the Radiotherapy and Cancer Residency Program conducted for physicians. A 2700-curie ^{60}Co facility (Fig. 4) in the basement of this building had a calculated exposure rate, neglecting shielding, self-absorption, and scattering, of 59 r/min at 1 m on Dec. 11, 1963. Another ^{60}Co irradiator is located in the PRNC animal house.

A 10,000-curie ^{137}Cs source (Fig. 5), developed by American Nuclear Corp., Oak Ridge, Tenn., has been used to irradiate a portion of the lower montane rain forest as part of the terrestrial ecology work.

Several plutonium–beryllium neutron sources and other small radiation sources are available for research use.

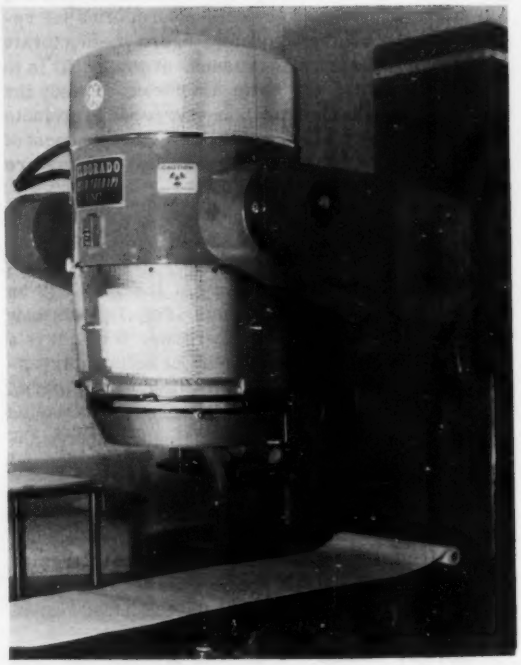


Fig. 3 The large medical teletherapy unit located in the PRNC Bio-Medical Building.

Research Program

Research is conducted at PRNC both in conjunction with its training objectives and under the direct sponsorship of the USAEC Divisions of Research, Isotopes Development, and Biology and Medicine. Much of this research is directed toward the solution of problems encountered in a tropical environment but possibly of vital significance both in the continental United States

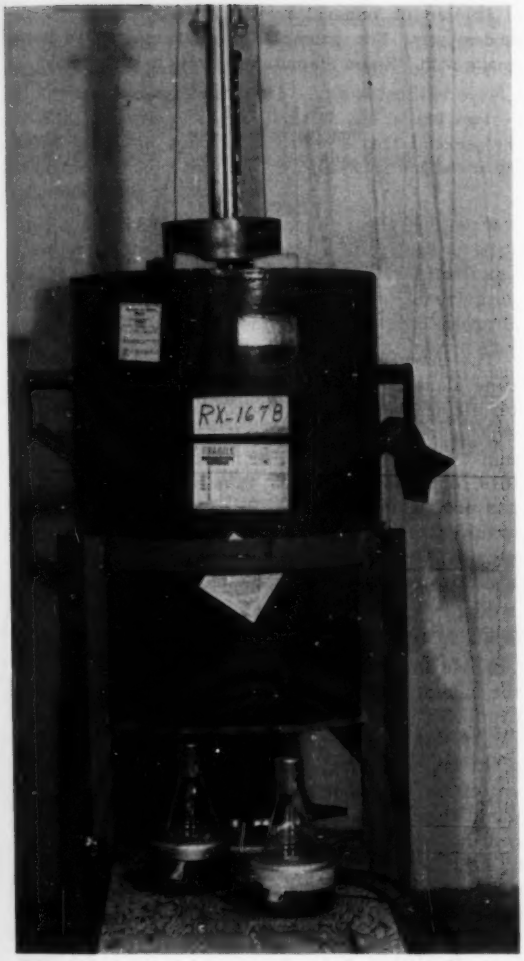


Fig. 4 The 2700-curie ^{60}Co gamma-irradiation facility in the basement of the PRNC Bio-Medical Building.

and elsewhere throughout the world. The programs described here are directly related to the use of isotopes. However, work is also being carried out on radiation damage in organic crystals and ferroelectrics, formation of color centers by irradiation of alkali halides, neutron diffraction in relation to chemical bonding, and resonance ion radiation in relation to radiation damage in biological systems.

RADIATION PRESERVATION OF FOODS

Several exotic tropical fruits have considerable market potential if their ripening can be delayed to permit shipment. The feasibility of radiation preservation of mangoes and bananas is being studied from two aspects: (1) determination of the preirradiation conditions, radiation dose, and postirradiation treatment that delay ripening and maximize the shelf life of the food product and (2) measurement, by appropriate biochemical assay, of changes in nutritional factors that accompany radiation pasteurization. Qualitative or semiquantitative criterions of ripening—softening, color change, spotting, and taste—are being determined, and characteristics associated with ripening, such as the decrease of starch and increase in sugar in bananas, have been measured. Biochemical study includes the quantitative assay of vitamins and the effect on them of radiation dose and treatment.

Experiments are being conducted with 20 varieties of mangoes growing on the PRNC grounds in Mayaguez. A 0.25-Mrad dose of gamma radiation has been found to extend the shelf life at 50°F approximately 20 days for some varieties. Measurement of the extent of depolymerization of the pectic constituents of mangoes during and after irradiation has shown softening of the fruit to be due to both radiation-induced depolymerization of pectic acids and to the considerable polygalacturanase activity in the fruit.

Fig. 5 The 10,000-curie ^{137}Cs source in the El Verde Rain Forest during the final stages of installation.

Experiments with Monte Cristo variety bananas from the UPR Agricultural Experiment Station plantation have shown doses between 20 and 40 kr best for retardation of ripening.

Assays for vitamins A and C as a function of time after irradiation are run routinely on all irradiated fruit, and results are correlated with the stage of ripening and radiation dose.

SUGARCANE MUTANT PRODUCTION

To increase the economic return from sugarcane, Puerto Rico's most important crop, the induction of plant mutants with high sucrose content is being attempted. The radiation sensitivity of seeds and buds to thermal neutrons from the 1-Mw reactor was determined, and thousands of seeds and vegetative buds have been irradiated, germinated, and planted in the field. The individual plants produced will be mass-screened for sugar content by automated analytical techniques. Visible mutations, such as wider and stiffer leaves, indicate that other favorable characteristics may be induced. Superior mutants will be propagated and evaluated in the UPR Agricultural Experiment Station's program of crop breeding and improvement.

A program on induction of sugarcane resistance to the mosaic virus disease is under way. Nearly a thousand plants have been grown from irradiated seeds and will be mass-screened by artificial infection with the virus.

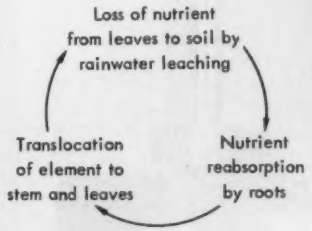
SUGARCANE BORER CONTROL

The sugarcane borer, *Diatraea saccharalis* (Fab.) (Crambidae: Lepidoptera), causes direct destruction of plant tissues, often completely destroying young plants as well as causing sugar inversion in the cane stalks. In addition, this pest allows secondary invasion by fungi and bacteria, which in turn decrease sucrose yields. It is cosmopolitan in distribution and of major importance in most sugar-producing areas of the world. The chief objectives of the research are determining whether the sterile-male technique, using gamma irradiation for sterilization, can be applied to eradication of this pest. The project is part of a cooperative effort with the UPR Agricultural Experiment Station.

TRACER STUDIES IN SUGARCANE

Important problems of both immediate and long-range application are being studied in sugarcane through the use of radioisotopes. Several field and greenhouse experiments, which deal with agronomic practice, are in progress. For example, the effect of soil factors, *p*H, density, and moisture content on uptake of nutrients and utilization of phosphates, sulfates, and trace elements is being determined. The results of studies on enhancement of foliar absorption of phosphates by wetting agents will be useful in the aerial spray application of fertilizers to sugarcane, an increasingly common practice.

Studies of mineral cycling in tropical crops are under way. For example, measurements are being made with ^{65}Zn on the nutrient cycle in sugarcane:



The enzymatic degradation of sucrose in the sugarcane plant by invertase has been investigated by determining factors that control invertase formation. For incorporation of added ^{14}C-labeled amino acids into the protein fraction of meristem tissue, cofactors, e.g., magnesium ion, have been found essential, and protein formation has been shown to be considerably decreased in the presence of sulfhydryl compounds. Nucleotides found in such tissue are being identified by chromatographic techniques.

RAIN-FOREST ECOLOGY

In one small area of montane rain forest, 1500 ft up the side of El Yunque in eastern Puerto Rico, studies are being made of (1) the effect of radiation on the rain-forest ecological system, one of the most complex of all natural systems; (2) mineral cycling in tropical vegetation with emphasis on fallout, earth moving, and waste disposal; and (3) basic functional features of the rain-forest system — metabolism transpiration, and biological circuits — needed to understand the phenomenons related to the first two objectives. A rain-forest field station is in operation, the forest has been irradiated, the basic annual cycle of biotic events has been established, basic dimensions of the metabolism have been measured, gamma spectrum studies are in progress, and chemical tracer experimentation has begun.

MARINE BIOLOGY

In an integrated research program designed to provide data on the distribution and movement of trace elements in a restricted, but complete, ecological and biogeochemical system, measurements are being made of (1) biological productivity, (2) distributions of selected trace elements in the system, (3) concentration of given radioisotopes by selected organisms, (4) distribution of worldwide fallout in marine organisms and sediments, and (5) physical and chemical oceanography.

Work is under way in an area on the west coast of Puerto Rico in which special emphasis is on food–web relations. The area under study includes the watersheds and offshore areas of three rivers, which

drain, respectively, watersheds predominantly comprising limestone, volcanic soil, and a soil with extensive outcroppings of serpentine. The three rivers empty into the sea within a 17-mile stretch of coast, and the amounts of trace elements and fallout radionuclides in the marine organisms that inhabit the estuarine areas of this region reflect the influence of the neighboring watersheds.

Study for Proposed Transisthmus Canal. A survey in estuarine and marine ecology is being made as part of a feasibility study, by Battelle Memorial Institute, for a sea-level isthmian canal in Central America. It includes analysis of currently available predictions of radionuclide production and distribution, interpretation of these in terms of the specific-activity approach to environmental contamination problems, and identification of the radionuclides that appear most likely to constitute a potential hazard to man through the estuarine and marine environments.

CANCER RESEARCH

Cancer studies include investigation of the role of surgical sterilization in the etiology of cancer of the uterine cervix, the fractionation of weekly radiation doses in cancer patients receiving radiation therapy, carcinoma of the uterine cervix associated with pregnancy, biochemical changes in the blood of cancer patients receiving radiation therapy for cancer of the esophagus, chromosome changes in patients receiving radiation therapy, lymphangiography in cancer patients, exfoliative cytology as a tool for determining prognosis in cases with cervical carcinoma submitted to irradiation, a surgical adjuvant breast project, and a study of the incidence of leukemia in patients with cervical cancer treated with radiation.

CLINICAL RADIOISOTOPE RESEARCH

Research is being conducted on diagnosis and therapy of thyroid disorders, gastrointestinal malabsorption, renal disorders, and organ and tumor localization.

CHEMISTRY

Hot-Atom Chemistry. Research in hot-atom chemistry concerns studies on aromatic organometallic compounds, including those of bismuth, indium, tin, mercury, thallium, phosphorus, and sulfur, which are irradiated as solid compounds and in benzene solution. Equipment has been constructed to extend this work to the gas phase and to short-half-life isotopes. The fundamental mechanism of the radiolysis of hydrogen chloride and hydrogen bromide is being studied, using both gamma radiation and fission recoil particles. This study is part of an overall investigation on the conversion of fission energy into electrical energy, using a cycle involving radiolysis of a hydrogen halide by fission fragments to provide feed for a fuel cell. Radiation-induced reactions of organic compounds in solution and the mechanism of thermal rearrangements are also being studied with isotopes.

Photochemistry and Radiation Chemistry. Advanced training is provided in photochemistry and radiation chemistry, with special emphasis on their relations. Research topics include studies of transient species formed by gamma irradiation of heterocyclic molecules and similar projects on transient species formed by photoionization and photodecomposition. The related theoretical projects involve molecular orbital and valence bond calculations for heterocyclic molecules. Gamma radiolysis and photolysis studies have recently been started. Molecular orbital calculations are being carried out, and a large number of heterocyclic molecules have already been evaluated. Valence bond calculations in triplet states of heterocyclic molecules are in progress.

Organic Chemistry. Advanced training in organic chemistry emphasizes its nuclear aspects. Research topics include the use of ^{35}S in exchange reactions and of ^{36}Cl and ^{14}C to determine reaction mechanisms, synthesis of boron compounds of potential use in neutron-capture therapy, and radiolysis of dimethyl sulfoxide. (MG)

Use of Radioisotope Techniques in Sweden

By Lars G. Erwall*

Abstract: Radiotracer work in Sweden is carried out under both government and private-industry sponsorship. As examples of the fields of activity, the uses of radiotracers in industrial processes, furnace-lining wear, and water pollution, in identifying metal products, and in activation analysis are discussed.

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The Isotope Techniques Laboratory (ITL) was organized in 1960 as the outgrowth of several years of research and development work on industrial isotope techniques in the Division of Physical and Nuclear Chemistry of the Royal Institute of Technology. ITL, which is supported by member companies, has maintained an extensive research and development program sponsored by the Government Research Councils

and private funds. It arranges courses on uses of radioisotopes and radiation sources in industry and of isotope techniques in analysis.[1] In the past it has performed commercial contractual work, about half of some 600 contracts received having been for radioactive tracer study of industrial processes and water pollution. Early in 1966 the contractual work was taken over by a subsidiary, AB Isotopteknik (IT), and ITL now only does research and disseminates information on isotope applications to its members.

Industrial Processes

Radiotracers have found extensive use in studies of industrial processes, especially in the pulp and paper industry.

Radioisotope-labeled chips have been used to follow wood chips through continuous pulp digesters. For small digesters an irradiated copper wire, containing ^{64}Cu, is inserted in a piece of wood,[2] and for large vessels irradiated Teflon chips, containing ^{24}Na, are introduced.[3] The retention time for the individual chips is determined from data recorded by detectors mounted at the inlet and outlet. For the mapping of the movement of the chips in the digester, detectors are mounted at different heights close to the wall. The maximum reading of the count rate for the detectors gives the passage time at the different levels. The absolute value of each of these readings gives the attenuation of the gamma radiation, and this is used for calculating the distance of the chip from the detectors at each level.

The mean residence time for cellulose fibers in a bleaching tower is found by adding irradiated glass fibers, containing ^{24}Na, to the incoming pulp and continuously measuring the activity at the outlet. The effective volume and the mixing properties of the vessel can also be evaluated from activity-vs.-time curves. Similar studies have been made on continuous mixing and residence time in vessels in the pulp and paper industry.

In metallurgical operations the movement of material through cupola and blast furnaces has been investigated with radioisotope tracers. For example, the retention times of scrap iron, limestone, and coke were found from measurements made on tracer ^{140}La or ^{198}Au that had been inserted in lumps of the material under study.[4] As a bonus the average residence time of molten steel in the furnace was established.

The efficiencies of mixers of different design[5] for lightweight concrete were compared by following the movement of radiotracers induced in the components — cement powder and alumina powder — by irradiation. In the cement ^{24}Na and ^{42}K are induced, and in the aluminum, ^{56}Mn from manganese impurity. Ready access to a reactor is important for such a study since the half-life of ^{56}Mn is only 2.6 hr.

Water Pollution

Accurate data on river flow rates are necessary for evaluating the hazards to organic life downstream in the river due to, for example, oxygen-consuming wastes. Because conventional flowmeters must be calibrated and are not always easily installed, radioisotope tracer methods have found frequent use, especially in investigations of waste-disposal systems. The velocity ("peak-timing") method is best suited for measurements of turbulent flow in closed conduits, where the flow fills the whole cross section of the conduit, whereas the constant-rate injection ("dilution") method is used in open channels and streams. Under favorable conditions an accuracy of 0.1 to 0.5% is attainable in practical operation with both methods for measuring liquid flow, which is as good as can be achieved by weighing the discharge.

The minimum velocity of the river water must be known for calculating the river's permissible load of industrial wastes. For determining the mean transport time between the point of discharge of the waste into the stream and a given downstream point, a tracer is simply injected at the actual or planned waste-discharge point and its activity is continuously recorded at the downstream point. The mean passage time is the time coordinate of the center of gravity of the activity-vs.-time curve.

Extensive radiotracer studies have been made on the flow pattern and dispersion in a large receiver around the mouth of pipes installed or planned for municipal sewage or industrial waste disposal. The initial dilution of the waste in the vicinity of the outlet, its direction of flow, and its dilution during transportation can be assessed with high sensitivity by using radioactive tracers, usually ^{82}Br as ammonium bromide. The tracer solution is injected, or released, momentarily or continuously at the location under study, and activities at different depths are measured by scintillation detectors immersed in the water.[6]

In the sound between Sweden and Denmark, ITL and the Danish Isotope Center have cooperated in extensive studies of sewage disposal planned for several cities.[7] Cruising boats equipped for automatic registration of radiotracer concentration, time, and location have obtained data from which both the degree of purification required for sewage before it is released and the best outlet point could be determined.

Furnace-Lining Wear

The wear rate of the lining of the Kaldo furnace, a Swedish invention for producing steel, has been quantitatively measured and the durability of various refractory materials compared by radioisotope tracer methods.[8] A radioactive material is inserted at different depths in the lining, and the activity of steel samples from each heat is measured. The half-life and radiation characteristics of ^{192}Ir make it a good

isotope for this purpose; it is used as an iridium–platinum alloy, which has a high melting point and a low affinity for oxygen.

Metal-Products Identification

The applicability of radioisotope labeling to routine identification of metals has been studied extensively,[9] and isotope labeling has proved to be a useful tool for positive identification of, for example, steel products of special runs. In a special steel plant[10] that made the heat-exchanger tubes for Sweden's first nuclear power reactor, a certificate of the complete chemical composition of each tube was required by the customer. Analyses of these tubes are very time- and labor-consuming. However, addition of a small amount of ^{182}Ta to the heat and measurement of the activity of the tubes made from it made it possible to identify each tube with the heat, whose chemical composition was already known. This nuclide was chosen because a rather long half-life was needed owing to the several months' span of the tube-manufacturing process. When an order was received later for tubes with somewhat different composition, ^{192}Ir was used because its gamma radiation is easily distinguished from that of ^{182}Ta. Tube activities were measured with a scintillation counter with a variable-level discriminator. When this was set to accept pulses corresponding to gamma energies above 0.01 Mev, all labeled tubes could be distinguished from unlabeled ones. When it was set to accept pulses corresponding to gamma energies above 0.65 Mev, only ^{182}Ta was recorded.

In the very simple case of labeling of one heat to avoid confusing ingots made from it with those from other heats, tracers with not too long half-lives and low radiotoxicity are usually used. In a more sophisticated application — simultaneous identification of several heats, each labeled with one identifiable nuclide or mixture of nuclides — reasonable amounts of radiotracers can be used, but expensive counting equipment and relatively long measuring times are needed.[10]

Activation Analysis

In the biocide residue debate in Sweden, attention has been focused on mercury because of the strong indication that a sharp decrease in the population of several species of birds of prey was caused by mercury poisoning. For acute poisoning, conventional methods can be used to determine mercury concentrations in the various organs, but a more sensitive method was needed to obtain a better picture of mercury distribution in chronic poisoning. Neutron activation analysis, which can measure mercury nondestructively down to about 0.1 ppm in biological samples and, if chemical separation is used, with a sensitivity more than 100 times as great,[11] was applied to a large number of bird and animal tissue samples. The results pointed to serious poisoning of several bird species whose dietary habits were such that the use of alkyl mercury compounds in agriculture was implicated. There was strong evidence[12] that a significant increase in the mercury content of bird feathers occurred simultaneously with the introduction of such compounds on the market for large-scale use. In this work, feathers from museum collections were examined and covered the period from around 1850 to today. The alkyl mercurials were therefore banned as agriculture fungicides, and less toxic compounds, e.g., alkoxyalkyl mercury compounds, were substituted.

However, agricultural mercurials could not be blamed for the high mercury contents found in certain other samples, e.g., fish. The research program was therefore broadened, and other sources of the mercury in wildlife are now being sought, together with the nature of the mercury compound in various tissue samples. More than 9000 samples have already been analyzed.

ITL has also cooperated in studying the occurrence of selenium in pigs[13] and in analysis of the remains of King Erik XIV, suspected to have been poisoned with arsenic by his brother in 1577, in order to find the cause of his death.[14]

Neutron activation analysis is also used for determining several trace elements in blood and other types of biological samples, and a rapid automated technique has been developed for chemical separation of different elements.[15,16] (MG)

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STABLE-ISOTOPE ENRICHMENT BY GAS CHROMATOGRAPHY*

Gas chromatography, which has been used successfully for enrichment of hydrogen isotopes, was recently shown by the Société National des Petroles d'Aquitaine to be applicable to carbon and neon isotopes, also. With Molecular Sieves 5A as the stationary phase and carbon monoxide as the gas phase, carbon isotope enrichments up to 9% were possible. A mass spectrometer was connected to the gas chromatography column in which the isotopes were separated to provide continuous and simultaneous analysis of the fractions. Neon isotope enrichments up to 7% were obtained with a similar setup except that in the mass spectrometer a single collector was used with magnetic sweeping and the two isotopes were determined simultaneously by a method based on integration of the signals. With argon isotopes, separation was very low. (Martha Gerrard)

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Section III

Isotopes and Radiation Technology

Process Radiation Development

Sixth Annual Contractors' Meeting on Process Radiation Development

By R. E. Greene

Abstract: Papers presented at the USAEC's Division of Isotopes Development Sixth Annual Contractors' Meeting, held at Germantown, Md., September 26–27, 1966, on process radiation development are briefly summarized. The papers covered work on (1) polymerization and copolymerization, (2) radiation-induced synthesis, (3) chemical reactors for radiation processing, (4) source development, (5) irradiation studies, and (6) radiation processing of materials.

The USAEC's Division of Isotopes Development (DID) held its sixth annual contractors' meeting on process radiation development at Germantown, Md., September 26–27, 1966. Martin H. Stein, in his opening remarks, stated that radiation processing is a multimillion dollar industry and is growing rapidly. He cited the dollar value of radiation-produced materials in the United States as $\$20 \times 10^6$ in 1963, $\$70 \times 10^6$ in 1964, $\$150 \times 10^6$ in 1965, and an estimated $\$250 \times 10^6$ in 1966. The bulk of the papers presented centered on radiation-induced polymerization, copolymerization, and other chemical reactions and syntheses. There were also papers on development of chemical reactors for radiation processing, neutron transmutation techniques for semiconductors, source development, dosimetry, comparison of ^{60}Co and ^{137}Cs, microbial inhibition, and other subjects. This review was prepared primarily from the program summaries distributed at the meeting. A formal report of the program summaries for 1966, similar to those for previous years,[1–4] has been published.* The paper numbers, titles, authors, and authors' affiliations are listed in Table 1.

Polymerization and Copolymerization

Steinberg (Paper 1) described the Brookhaven National Laboratory (BNL) Radiation Processing Program for investigating the use of isotopic energy for the synthesis and production of industrial chemicals. The relatively high unit cost of isotopic energy tends to restrict chemical processing to high-G-value or chain reactions, i.e., polymerization, oxidation, halogenation, and catalysis. For this program, survey experiments are performed with capsules in a radiation field to determine yields and types of products formed; reactions are studied to determine process chemistry, reaction kinetics, and product characteristics; and radiation chemical processes are designed and evaluated from the survey and advanced investigations. Specific systems under investigation are:

1. Homopolymerization of ethylene. Design and evaluation studies indicate promising economic potential. A small-scale flow experiment has been constructed to provide data and sufficient quantities of polymer for evaluation by industry (Paper 2).

2. Copolymerization of ethylene. Experiments indicate efficient ^{60}Co gamma-radiation-induced copolymerization of ethylene with other materials—for example, CO and SO_2, which are attractive because of their low cost. A study of the ethylene–CO system has been completed, and the study of the ethylene–SO_2 system is in progress (Paper 3).

3. Plastic-impregnated concrete. The use of ^{60}Co to polymerize a monomer that has been absorbed in the pores of concrete is being studied. The purpose of the process is improvement of the corrosion resistance of concrete that may be used in construction of distillation units in desalination plants.

4. Wood–polyethylene combination. Irradiation of wood in an ethylene atmosphere at 680 atm and 150°C with a 1.3×10^6-rad dose resulted in a 40% loading of polyethylene in the wood.

5. Synthesis of ammonia. Ammonia synthesis is an economically important industrial catalytic process, and any possible process improvements with the use of radiation should be determined. A definitive study on the effect of ^{60}Co gamma radiation on the catalytic synthesis of ammonia has been done (described by Dietz et al., Paper 10).

Kukacka (Paper 2) described the BNL investigation on using ^{60}Co gamma radiation to polymerize ethylene

*See Ref. 10, added in proof.

Table 1 PAPERS PRESENTED AT SIXTH ANNUAL CONTRACTORS' MEETING ON PROCESS RADIATION DEVELOPMENT

No.	Title	Author	Affiliation
1	Radiation Processing Program at Brookhaven National Laboratory	M. Steinberg, L. E. Kukacka, P. Colombo, R. Dietz, and B. Manowitz	Brookhaven National Lab., Upton, N. Y.
2	Engineering-Scale Studies on the Radiation-Induced Polymerization of Ethylene	L. E. Kukacka, P. Colombo, and M. Steinberg	Brookhaven National Lab., Upton, N. Y.
3	Radiation-Induced Synthesis of Ethylene Polysulfones	P. Colombo, J. Fontana, and M. Steinberg	Brookhaven National Lab., Upton, N. Y.
4	The Radiation-Induced Polymerization and Copolymerization of Unusual Monomers	V. T. Stannett, J. E. Herz, J. J. Kearney, and J. Cleaveland	Research Triangle Inst., Durham, N. C.
5	Radiation-Induced Emulsion Polymerization	V. T. Stannett, J. A. Gervasi, and J. J. Kearney	Research Triangle Inst., Durham, N. C.
6	The Effects of Ionizing Radiation on Aqueous Solutions of Water Soluble Polymers	H. R. Spragg	Unidynamics/Phoenix, Phoenix, Ariz.
7	Radiation-Induced Codimerization of Ether-Unsaturate Mixtures	C. D. Bopp and W. W. Parkinson	Oak Ridge National Lab., Oak Ridge, Tenn.
8	The Search for Ionic Chain Radiation Reactions	W. H. Johnston	Johnston Laboratories, Inc., Baltimore, Md.
9	The Radiation-Induced Addition of Methylsilane and Dimethylsilane to Ethylene. A Gas Phase Ionic Reaction	W. H. Johnston	Johnston Laboratories, Inc., Baltimore, Md.
10	Heterogeneous Radiation Chemistry	R. N. Dietz and M. Steinberg	Brookhaven National Lab., Upton, N. Y.
11	Radiation Chemical Reactor Design Studies	F. B. Hill	Brookhaven National Lab., Upton, N. Y.
12	Development of a Radioactive Pebble Bed Chemical Reactor System	(Given by C. C. Thomas)	Western New York Nuclear Research Center, Inc., Buffalo, N. Y.
13	Source Development	J. I. Wagner, A. Oltmann, and L. Galanter	Brookhaven National Lab., Upton, N. Y.
14	The Operations Program of the High Intensity Radiation Development Laboratory	J. S. Milau and P. T. Fallon	Brookhaven National Lab., Upton, N. Y.
15	Temperature Distribution in Targets	P. T. Fallon and M. A. Rothbart	Brookhaven National Lab., Upton, N. Y.
16	Cesium-137, Cobalt-60 Comparison	F. X. Rizzo and L. Galanter	Brookhaven National Lab., Upton, N. Y.
17	Dosimetry	F. X. Rizzo and L. Galanter	Brookhaven National Lab., Upton, N. Y.
18	Calculation of Beta Particle Slowing Down	M. S. Zucker and H. Lichtenstein	Brookhaven National Lab., Upton, N. Y., and Columbia Univ., New York, N. Y.
19	Semiconductor Device Fabrication by Neutron Transmutation Techniques	C. N. Klahr	Fundamental Methods Associates, Inc., New York, N. Y.
20	Survey of Potential Microbial Inhibition Systems Employing Radiation (MISER)	J. Kivel, M. S. Chatterton, and R. J. F. Palchak	Atlantic Research Corp., Alexandria, Va.
21	Nuclear Radiation Processing of Optical Maser Materials	H. Sadowski, J. G. Beckerley, and K. Held	Radioptics, Inc., Plainview, L. I., N. Y.
22	Radiation Processed Wood Plastic Combinations	J. A. Kent and W. R. Boyle	West Virginia Univ., Morgantown, W. Va.
23	An Investigation of Certain Physical and Mechanical Properties of Wood–Plastic Combinations	R. L. Ely, Jr., and J. A. Merrill	Research Triangle Inst., Durham, N. C.

and to acquire radiation, chemical, and engineering data for the use of radiation as a standard unit operation in high-pressure systems. Results indicate that a wide variety of commercial-grade polyethylenes could be produced at high polymerization rates. Preliminary cost estimates for processes operating above and below the polymer melting point indicate conversion costs that may give the process economic potential. A small-scale flow system has been constructed, several hundred grams of polymer per run are being made, and characterization of the polyethylene is under way.

Radiation-induced synthesis of ethylene polysulfones was described by Colombo (Paper 3) of BNL. In this experimental program, Colombo et al. studied the kinetics for the gas-phase reaction first and for the liquid-phase reaction second. In gas-phase reactions carried out at pressures from 34 to 680 atm and temperatures from 20 to 60°C, copolymers of ethylene and SO_2 were formed from mixtures containing initially 0.1 to 26% SO_2. By varying the SO_2 concentration in the gas mixture, they were able to produce copolymers containing from 9 to 46% SO_2.

Liquid-phase experiments, with ethylene pressures from 34 to 680 atm and temperatures from 20 to 160°C, produced copolymers that had equimolar concentrations of ethylene and SO_2. The significance of the gas-phase research is that polysulfones with different ethylene concentrations and a wide range of properties can be produced by radiation. The raw materials are cheap, and the polymerization rates are much higher than those for the radiation-induced homopolymerization of ethylene under similar conditions.

Stannett (Paper 4) described work at the Research Triangle Institute, Durham, N. C., on radiation-

induced polymerization and copolymerization of unusual monomers. Solid-state polymerization of trithiane, highly purified by two or more sublimations, could not be initiated by radiation. Acidic compounds produced by radiolysis of impurities probably initiated the polymerizations previously observed. Attempts to copolymerize trithiane with dithiane and trioxane were unsuccessful. An improved technique for the preparation and handling of relatively pure formaldehyde was a significant development last year. In low-temperature irradiations of vinyl ethyl ether, butadiene, isoprene, isobutene, *p*-chlorostyrene, or *p*-methoxystyrene mixed with liquid formaldehyde, G (monomer) values varied greatly, but some were as large as 3×10^5. Analysis of some of these mixtures showed the products to be predominantly polyformaldehyde. Comonomer units, if present at all, were less than 3%. Low-temperature irradiations of sulfur dioxide–diene mixtures with a third monomer have continued, and terpolymers with methyl methacrylate and trifluorovinyl acetate have been obtained.

Stannett (Paper 5) also described studies on radiation-induced emulsion polymerization of vinyl acetate which included kinetic studies at 0, 30, and 60°C and effects of temperature, radiation intensity, and emulsifier concentration on molecular weight, extent of branching, and latex particle size and properties. Copolymerization of vinyl acetate with various maleate and acrylate esters is also being studied relative to polymerization rate, copolymer molecular weight, and latex properties. Emulsion polymerization of vinyl acetate has been shown to deviate from the classical Smith–Ewart theory of emulsion polymerization in nearly every respect.

The paper by Spragg (Paper 6), of Unidynamics, Phoenix, Ariz., on the effects of ionizing radiation on aqueous solutions of water-soluble polymers was given by R. L. Curtin. Ultimate goals of the present program are the advancement of the science of colloid systems and disclosure of radiation processes of potential industrial significance. Work has included the study of the effects of ^{60}Co radiation on the rheology and chemical nature of aqueous sols of polyvinyl alcohol, acrylamide, and polyoxyethylene. Upon irradiation of solutions of high-molecular-weight polyoxyethylene, there is a loss of viscosity at low doses followed by abrupt gelation at total doses of 2 to 5×10^4 rads. The versatility of ionizing radiation for the control of viscosity in solutions without addition of extraneous chemicals has been repeatedly demonstrated. Such systems as oil emulsions or suspensions of rather large particulate matter may be stabilized with polyethylene oxide to promote a chemical reaction. After reaction, the components may be separated by the reduction of the viscosity that results from a dose of a few thousand rads. Initial results from the preparation of acrylamide gels have prompted a study of this polymerization because of its possible use in technical and industrial applications. With controlled application of modest doses and dose rates, the viscosity of the solution can be increased at will, the resulting heat of polymerization can be used to produce stepwise (incremental) increases in the temperature of the system, and the dose rate can be monitored from the slope of the temperature–time curve.

Radiation-Induced Synthesis

Bopp (Paper 7) of Oak Ridge National Laboratory discussed radiation-induced codimerization of ethers with unsaturated compounds. Useful products may possibly be produced in high yield by this method. Solutions of cyclohexene or dihydrofuran in a saturated furan derivative were irradiated with a ^{60}Co source, and codimer and dimer yields were determined by gas chromatography. Although yields of certain of the products increased at higher temperatures, very high yields of codimers were not achieved; however, other systems using more reactive olefins are expected to give higher yields at higher temperatures.

Wm. H. Johnston of Johnston Laboratories, Inc., Baltimore, Md., gave two papers (Papers 8 and 9) on ionic chain reactions. Such reactions would offer a significant contribution to an area of chemistry that could be unique to radiation processing. A gas-ion chain reaction has been discovered in the radiation-induced additions of methylsilane and dimethylsilane to ethylene. In thioolefin studies the chain reaction appears to be solely of a free-radical nature. Gamma irradiation of a methyl mercaptan–ethylene mixture formed a single product, $CH_3SCH_2CH_3$, with G values up to approximately 7×10^3. In the alcohol–olefin system, evidence was obtained for a theoretical prediction of an ionic reaction path, which can be expressed in terms of a generalized acid-catalysis theory.

Dietz et al. (Paper 10), BNL, in work on heterogeneous radiation chemistry, have been studying two effects of radiation on a solid substance in the presence of a gas: (1) the change in the electron configuration on the active sites on the surface and (2) transfer of energy from the surface of the substrate to the material adsorbed on the surface. Two approaches have been used to study these effects, respectively: (1) the synthesis of ammonia using a commercial iron catalyst and (2) the decomposition of CO_2 adsorbed on various oxide semiconductors, e.g., Al_2O_3. Ammonia synthesis in the presence of a 9.5×10^5-rad/hr source showed no significant difference in yield from that without radiation. With a higher intensity radiation source, $\sim 4.4 \times 10^6$ rad/hr, an unexpected, irreversible decrease in the catalyst activity was apparent from the decrease in the ammonia yields. The effects of O_2 impurity in the synthesis gas will be studied, as will the irreversible

decrease in catalyst effectiveness with higher intensity sources. From the study of the decomposition of CO_2 in Pyrex reaction vessels, it is concluded that the decomposition rate depends on the type of semiconductor—insulator, *n*, or *p*—and its degree. Energy appears to be transferred from the surface of the solid to the gas adsorbed by the monolayer; and the presence of adsorbed hydroxyl groups causes a reaction other than strict decomposition, i.e., the intermediate formation of an adsorbed molecule similar to formic acid which then decomposes to CO_2 and H_2O. Increasing the irradiation temperature when η-Al_2O_3 is used as the solid sorber increases the decomposition rate up to 180°C; above this temperature the rate decreases because the O_2 product is desorbed and converted back to CO_2.

Chemical Reactors for Radiation Processing

Hill (Paper 11), BNL, described studies for the design of chemical reactors for radiation-induced processes. Work has been concerned with the relation of the kinetics of photoinitiated reactions to physical rate processes of fluid dynamics and heat and mass transfer. Calculations of chemical-reactor performance are being made, and variables such as overall reaction rate, yield, and molecular-weight distribution are being calculated. The effects of reaction mechanisms, reactor geometry, residence-time distribution, and heat release rates are sought. Expressions derived from these calculations are often useful for design purposes. Experiments to test models of reactor performance and to measure parameters of the models have been made using ultraviolet light to initiate the reaction. Important and basic theoretical foundations have been laid for the study of effects of diffusion, mixing, and residence-time distribution on nonuniformly initiated homogeneous reactions, especially photoreactions. Results from theoretical work plus those from experiments now in progress will provide the experimenter and the designer with necessary guidance.

Thomas (Paper 12) of Western New York Nuclear Research Center, Inc., Buffalo, N. Y., reviewed the experimental design, construction, and demonstration of a pebble-bed catalyst system—a chemical-reaction vessel containing a very large number of tiny radioactive beads. A mixture of liquids to undergo chemical reaction flows through the reactor and is intimately exposed to the radiation to initiate the reaction for conversion to the desired product. The system under consideration is one for chlorination of toluene, and both addition and substitution products are formed. By controlling reaction temperatures, it should be possible to make the substitution reaction, giving benzotrichloride, predominate. In the first phase of the work, alpha, beta, and gamma sources were studied and economic and engineering evaluations made. Results are being used to design bench-scale experiments which will demonstrate the feasibility of the radiation process. Calculations of process parameters, fluidization velocity, reactor length and diameter, and mass of pebbles have been completed for both laboratory- and production-scale equipment. The pebble-bed system should permit more efficient use of emitted radiation, thus decreasing source investment.

Source Development

The prime objective of the BNL source-development program, according to Galanter (Paper 13), is the production of isotope forms best suited as radiation sources for industrial processing or for special uses. Methods have been developed for calculating self-absorption in rectangular and cylindrical sources, dose-rate distribution from ^{137}Cs within homogeneous targets, and specific dose rates for comparing ^{137}Cs sources of various thicknesses. Work with ^{60}Co sources includes new fabrication methods, nondestructive tests, and corrosion studies. Future research on sources will center on lower source cost, design calculations for ^{137}Cs sources, high-temperature stability, and safety.

Irradiation Studies

The operations program of the High Intensity Radiation Development Laboratory (HIRDL)[5] at BNL was described by J. H. Cusack for Milau and Fallon (Paper 14). Experiments include studies on heat generation in gamma-irradiated targets, comparison of ^{137}Cs and ^{60}Co, corrosion in conjunction with the source-development program, calorimetric determination of source activities, and dose distribution in various targets. Included with the paper was an interesting color film showing how 30 thousand lb of bacon was irradiated for the U. S. Army Quartermaster Corps. The HIRDL source plaque activity was increased to over 8×10^5 curies, and the bacon was given a minimum dose of 4.5×10^6 rads, the maximum allowable dose being 5.6×10^6 rads. Two extracorporeal ^{137}Cs blood irradiators of 4 thousand curies each have been fabricated at BNL. Other work included reviewing designs, testing irradiators, and encapsulating 140 thousand curies of ^{60}Co for various users. About 2800 separate irradiations were performed during the past year.

Galanter also presented a paper for Fallon and Rothbart (Paper 15) of BNL on a study of heat generation in targets exposed to large radiation sources. In addition to experimental work on energy flow, mechanisms for disposition of this energy, and the position at which maximum temperature occurs, a theoretical analysis of the problem is also being made. The thermal conductivity of bulk quantities of materials such as grain is so low that removal of absorbed energy by conduction is impossible. Even in

materials of high thermal conductivity, the energy from higher intensity sources is deposited so rapidly that only a small portion of it can be removed during irradiation. Greater knowledge of the transient thermal conditions existing in targets will be of value in the designing of future irradiators.

Rizzo (Paper 16) described work at BNL comparing depth-dose distribution of ^{60}Co and ^{137}Cs using homogeneous and heterogeneous targets of various sizes, shapes, densities, and atomic numbers. Preliminary results indicate that the depth-dose distribution curve from ^{137}Cs is flatter than that from ^{60}Co for targets of low densities; therefore, for target materials of low densities, irradiation with ^{137}Cs may be advantageous. Data and techniques for the rigorous calculation of dose distributions in heterogeneous target systems are virtually nonexistent; this information is needed in the design of irradiators and should be available for ^{137}Cs as well as ^{60}Co. In the discussion that followed, Galanter emphasized that in comparing sources, each specific application must be considered in itself.

Rizzo (Paper 17) also discussed the BNL dosimetry program, which is directed toward the development of new dosimeters; the evaluation, standardization, and improvement of techniques and procedures for some of the older systems; and the dissemination of dosimetry information. Liquid-, solid-, and gas-phase dosimeters have been considered, with sensitivities ranging from 10^3 to 10^7 rads and target-material densities from < 0.1 to 2.5 g/cm^3. Development work has been completed on silicon-solar-cell, deaerated-Fricke, flow-chemical, UVT (Ultraviolet Transmitting)-Lucite, and miniaturized-Fricke dosimeters. In addition to studies of the isochronal and isothermal bleaching characteristics of the UVT-Lucite and new blue-cellophane dosimeters, the latter was further evaluated for linearity, reproducibility, and method of readout. Rizzo stressed that, when dosimetry measurements are made inside the absorbing medium, the dosimeter should be of the correct size, shape, density, and atomic number relative to the medium in which it is used. The response of each dosimetry system should be thoroughly understood.

Lichtenstein (Paper 18), Columbia University, N. Y., described studies being made of the slowing down of beta particles in matter. The geometry assumed was that of the cone-shaped flux from a collimated point source incident on a slab of finite thickness and infinite lateral extension. Three objectives of this study were to determine (1) the energy spectrum and angular distribution of the beta particles and the deposited energy as a function of radial and axial position in the medium and in the two external regions; (2) the degree of accuracy with which a mass absorption coefficient to represent beta absorption in terms of the beta end-point energy can be determined; and (3) the effects of an improvement in the realism of charged-particle (particularly beta) energy-deposition calculations. A theoretical calculation of the Monte Carlo type was used, specially modified to take into account the peculiarities of charged-particle behavior in the slowing-down process.

Radiation Processing of Materials

Semiconductor-device fabrication by neutron transmutation techniques was described by Klahr (Paper 19) of Fundamental Methods Associates, Inc., N. Y. Results achieved in this program include fabrication of field-effect transistors of acceptable quality, and resistors, diodes, and capacitors of silicon and germanium that have a variety of characteristics; demonstration of the use of a surface oxide layer and its associated inversion region in transmutation-doped devices and of the practicality of thin—2 mils or less—cadmium irradiation dies (use of dies permits practical control to within 1 to 2 mils, while the use of an oxide surface layer permits control to within 1000 A normal to the surface); and partial fabrication of simple microcircuits. Techniques have been developed to carry out the transmutation doping process used in irradiation die materials by bonding a thin layer of cadmium to a thicker layer of a metal transparent to thermal neutrons, e.g., copper or steel; photoetching of the irradiation die; enclosing the die and semiconductor specimens in a suitable irradiation capsule; specifying the neutron flux and the semiconductor resistivities to ensure the doping of the spatial regions, i.e., to ensure that $p-n$ junctions are formed; and adapting various standard semiconductor techniques to these materials after the transmutation doping to fabricate electronic devices. The important results of this work appear in the controllability and predictability of the doping. The structure of the product was that of a SFET (Surface Field Effect Transistor), which appears to be the most favorable transistor structure for transmutation doping. Resistors in the range of 5 to 100 thousand ohms have also been produced by the same techniques. Hence transmutation doping has now produced all the components of microcircuitry—transistors, diodes, resistors, and capacitors—in single-crystal chips, and the process is ready for use in fabrication of complex silicon and germanium microcircuits.

Transmutation doping has three potential areas of advantage for microcircuitry fabrication: (1) performance characteristics not obtainable by other processes (diffusion or epitaxy); (2) potential low cost for either few- or many-of-a-kind production; and (3) potentially greater controllability of fabrication. The unique geometric configurations that can be produced in transmutation-doped devices impart performance advantages because

1. The doped regions can be of very uniform concentration.

2. Clear-through doping can be achieved, which permits interconnections on the bottom of the slice and allows an increase in the number of devices on each slice.

3. The $p-n$ junctions can be formed either perpendicular to the semiconductor surface by clear-through doping or parallel to the surface at any desired depth.

4. Junctions with desired concentration gradients can be formed in either high- or low-resistivity material.

5. Junctions with large reverse breakdown voltage and large capacitance change can be formed by appropriate choice of concentration gradient.

6. Intricate three-dimensional doping patterns can be produced with appropriate radiation die designs.

These configuration advantages lead to advantages in transistor and diode characteristics. We can expect capabilities in fabrication of diodes and transistors with a wide range of input impedance, gain, and rectification characteristics; resistors with a wide range of resistance values and condensers with a wide range of capacitances; new modes of interconnection of devices permitted by the two-sided configuration; and, in a single irradiation, microcircuits composed of many individual components within a single-crystal chip with all the advantages in electronic characteristics and interconnection capabilities of assemblies of individual transmutation-doped components. Several simple microcircuits have been made; however, the transmutation doping process has not yet been used to fabricate microcircuits with all the functional capabilities now existing in the semiconductor industry. Earlier work has been described in *Nucleonics*[6] and *Isotopes and Radiation Technology*.[7]

Kivel (Paper 20), Atlantic Research Corporation, Alexandria, Va., discussed a survey program to determine potential applications of processing systems using radiation for inhibiting biological and microbiological growth in nonfood materials. Studies had shown[8] that microbial growth in jet fuel can be arrested by relatively low doses of radiation. Other applications appeared promising. A literature review, equation derivation, and a detailed survey of industry, government agencies, armed forces, etc., constitute the three phases of the work. The large number of literature references obtained has been used to compile tables of radiation resistances of specific microorganisms, and criteria on the effect of radiation on microbial systems have been summarized. These criteria have provided a basis for equations to determine the amount of radiation required to control microbial growth in many systems. In the survey discussions, detailed information was obtained on parameters on specific systems, and these parameters have been fed into the equations to obtain preliminary estimates to determine whether or not irradiation costs are in the range of those for existing control methods. Estimates have been based on using BNL standard ^{60}Co strips; however, the selection of isotopes and use of ideal irradiator design may considerably reduce these preliminary estimates. Radiation inhibition of microbial growth in jet fuels, organic fuel holding tanks, machine cutting fluids, and in certain applications of cosmetic production appear to be economically feasible.

Sadowski (Paper 21), Radioptics, Inc., Plainview, L. I., N. Y., discussed research on how nuclear radiation can be used to create new materials for solid-state optical masers. It has been necessary to understand how radiation affects the optical properties of solids, to perform irradiation experiments on materials whose optical maser properties are substantially improved by radiation, and to make beneficial effects irreversible, i.e., eliminate thermal or optical bleaching. Laser rods of ruby, neodymium-doped barium silicate glass, and neodymium-doped soda lime–silicate glass have been irradiated with a 2.36×10^7-rad dose from ^{60}Co. The radiolytic reduction of elements in the neodymium glasses, added to facilitate their manufacture and not present in earlier samples, caused an unexpected result—the laser rods became totally opaque! However, controlled optical bleaching and thermal annealing caused both the barium silicate glass and the ruby to surpass their preirradiation laser performance. With maximum optical pumping energy, the maximum improvement in absolute efficiency was 25% with the neodymium-doped barium silicate glass.

Wood–Plastic Combinations

Kent (Paper 22) divided the work completed the past year at West Virginia University, Morgantown, W. Va., on wood–plastic combinations (WPC) into three areas: radiation polymerization of vinyl monomers, engineering support, and evaluation. Radiation requirements were determined for (1) completely polymerizing certain monomers both in bulk and in wood; (2) polymerizing monomers containing additives to promote cross-linking and plasticizing; and (3) polymerizing monomers containing inert additives to reduce heat generation and cost (diluents) or to enhance the quality of the product. Dose-rate studies are also being made. Engineering support involves work on uniform and nonuniform (shell type) impregnation of the wood, heat-transfer analysis relative to the use of those containers recommended in the Vitro Engineering pilot-plant study,[9] and planning experimental work for the new irradiation facility. Evaluation includes preparation of samples for testing by the Research Triangle Institute, examination of the microscopic distribution of polymer in the wood, and the study of diffusion of water and water vapor through the product. Work in progress is yielding a better understanding of the properties of the

material, and new systems with somewhat different properties are being developed.

Ely (Paper 23), Research Triangle Institute, discussed the investigation of certain physical and mechanical properties of WPC. The program is designed to provide a more significant quantitative evaluation of the mechanical properties of WPC, and since, for the most part, standard ASTM tests will be used, comparison of physical properties with those of other materials becomes possible. North Carolina State University is procuring, selecting, and preparing the wood test specimens and will perform the testing. West Virginia University is providing the monomer impregnation and polymerization services, and Research Triangle Institute has designed the experiments and will analyze the data. Four species of wood, four monomer formulations, and four loading (impregnation) levels for each formulation will be used. In addition, four logs have been obtained for each wood species. With multiple specimens required for each of several tests, 5120 tests will be required for the program. In addition to the primary objective of assessing changes relative to monomer loading, this program will also supply some qualitative information on the following sources of variation: differences between two trees of the same species, between vertical positions in the same tree, and between homogeneous groups (replication) within logs; tree-to-monomer loading and log-height-to-monomer loading interaction. Because of severe surface checking of some of the red oak and subsequent long drying time for new samples, only two monomers will be tested with this wood. Preliminary work with heavily loaded woods indicates that some changes in the hardness test methods will be required.

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Radiation Processing: Where We Stand Now

By David E. Harmer*

Abstract: Surveys of published literature indicate that a large number of radiation processes are either commercial or are in advanced development stages. It may be concluded that a radiation process industry is really operating, even if on a rather tentative scale. It may also be extrapolated that the known processes signify others which have not yet been made public but which may soon serve to broaden the scope of radiation as a processing tool.

The status and prospects for radiation processing were reviewed in a paper presented at the 1964 Conference on the Peaceful Uses of Atomic Energy.[1] It is now instructive to reexamine the potential of radiation processing and to attempt to answer questions concerning the rate of progress in this field. Since the area of food processing has been adequately reviewed recently,[2] this discussion is limited to other forms of radiation processing.

Wherever possible, accounts of processes either in advanced development stages or in actual commercial production have been taken from the published literature. The individual accounts are summarized in the accompanying tables, and the references can be used for obtaining detailed information on any given system. The general overview of this published literature is very similar to that in the earlier article. However, a number of proposed processes have now been commercialized, and a few new items have appeared. The growth of radiation as a commercial tool (Fig. 1), when compared with that of normal, established industries, is indeed remarkable.

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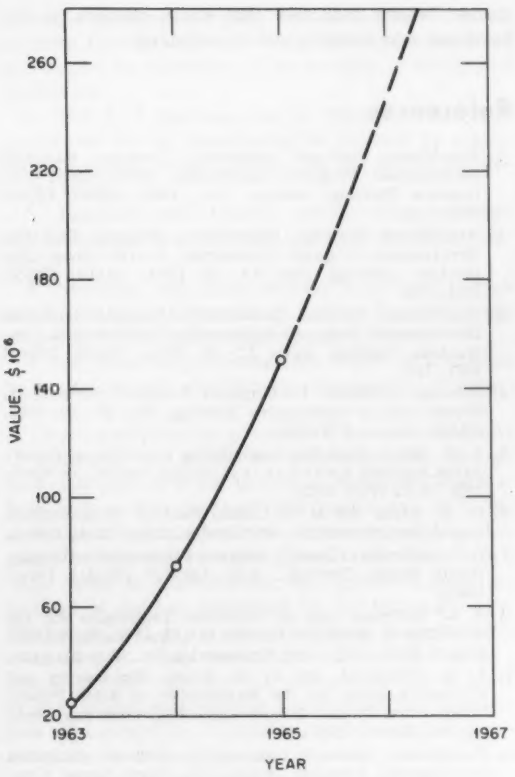

Fig. 1 Value of irradiated products in the United States during recent years, exclusive of food products.[3] This does not include the value of materials processed in industrial gamma sterilization plants of British design, which exceeded $56 million in 1965.

Radiation Sources

One of the most exciting developments of the last 2 years is the announcement[4] that ^{137}Cs was to be available at prices as low as 12.5 cents per curie in 200,000-curie quantities. Because the delivered energy per curie of ^{137}Cs is roughly 25% of that from ^{60}Co (50 cents per curie in 200,000-curie lots, 15 to 30 curies/g), such a price schedule now makes cesium extremely competitive. On the other hand, ^{60}Co is increasing its competitive position with the announcement[5] that large-scale quantities are about to be produced by a private organization in a private nuclear reactor.

The fate of ^{24}Na as a source of gamma radiation has been considerably less fortunate. The reactor at Hallam, Nebr., which had as one of its features a sodium-recycle system from which the ^{24}Na 2.75-Mev gamma rays were available, has been shut down and is being dismantled.[6] All indications are that no sizable quantities of chemical materials were ever processed by this sodium irradiator. While the reactor itself was shut down because of failures in other parts of the system, it should be noted that the location of this irradiator was extremely remote from any of the laboratories known to be leading in the field of applied radiation chemistry. This fact may account in part for the notable lack of success of the irradiator.

The trend in electron accelerators appears to be in the direction of a new generation of machines specifically tailored to meet the needs of a given product. One established manufacturer has announced a machine designed to answer needs in the area of thin, radiation-cured coatings.[7] A low-voltage accelerator has been developed by another company specifically for its own uses, but it is probably to be marketed through an instrument manufacturer new to the accelerator field.[8] The Insulating Core Transformer type of accelerator also appears to be making inroads into areas of potential commercial application.[9]

Sterilization of Medical Supplies

In terms of sheer quantities of radioisotopes being used, the field of sterilization of a variety of medical supplies[10–13] has a considerable lead over chemical and polymer processing. At present this application is growing at such a rate that even to maintain an accurate listing of all such facilities is a problem. Table 1, which is as up-to-date as possible, illustrates the present status.

Processing of Chemicals

Progress in processing of chemicals, as far as announced operating processes are concerned, has been disappointingly slow. Table 2 presents data for known processes that are either industrial or have been advanced to a late stage of development. The Dow ethyl bromide process, which has operated under irradiation for nearly 4 years, continues to be a very successful application. The original source, which at its maximum consisted of 2506 curies of ^{60}Co, was replaced in October 1966 with a new source of 3132 curies of ^{60}Co with a specific activity of about 10 curies/g. The new source restored this process to operating levels at least as great as those originally designed.

It is interesting that the sulfoxidation of hydrocarbons, a process that leads to biodegradable detergents, has now been carried out not only by the originator at Esso Research in Linden, N. J., but also by Farbwerke Hoechst in Frankfurt, West Germany. Both groups have done experimentation on a pilot-plant scale.

A process reported to have been carried out on an industrial scale is the oxidation of paraffins.[18,19] The procedure is especially interesting, since the stream

Table 1 COMMERCIAL IRRADIATORS FOR STERILIZATION OF NONFOOD PRODUCTS*

Product	Operator (designer)	Location (startup)	Radiation source	Capacity (load), curies
Goat hair and medical supplies	Gamma Sterilization Pty. Ltd. (AERE, Eng. Div.)	Dandenong, Victoria, Australia (1960)	^{60}Co	2,000,000 (665,000)
Disposable syringes	Johnson's Ethical Plastics, Ltd. (H. S. Marsh, Ltd.)	Slough, U. K. England (1961)	^{60}Co	300,000 (190,000)
Sutures	Ethicon, Ltd. (Nuclear Chemical Plant Ltd.)	Edinburgh, Scotland (1962)	^{60}Co	250,000 (82,000)
Medical supplies	Ethicon, Inc. (AECL)	Somerville, N. J. (1964)	^{60}Co	750,000–1,500,000 (480,000)
Medical supplies	Ethicon, Inc. (AECL)	San Angelo, Tex. (1964)	^{60}Co	750,000–1,500,000 (85,000)
Disposable syringes	Gillette Industries (H. S. Marsh, Ltd.)	Reading, U. K. England (1964)	^{60}Co	300,000 (75,000)
Medical supplies	Ethicon Sutures, Ltd. (AECL)	Peterborough, Ont. (1964)	^{60}Co	100,000 (35,000)
Medical accessories and food	Conservative Industries, "IRMA 400/1" (Saint-Gobain Techniques Nouvelles Co.)	Saclay & Lyon, France mobile (truck) (1965)	^{137}Cs	400,000 (173,000)
Scalpel blades	Swann-Morton, Ltd.	Sheffield, U. K. (1966)	^{60}Co	64,000
Medical supplies	Ethicon GmbH. (H. S. Marsh, Ltd.)	Hamburg, W. Germany (1966)	^{60}Co	750,000 (60,000)
Sutures	Braun Co. (Sulzer Brothers)	Melsungen, W. Germany (1966)	^{60}Co	600,000 (unknown)
Medical supplies	Tasman Vaccine Laboratory Ltd. (AECL)	Upper Hutt, New Zealand (1966)	^{60}Co	1,000,000 (160,000)
Disposable syringes	ICoSpa (H. S. Marsh, Ltd.)	Bologna, Italy (1967)	^{60}Co	500,000 (22,000)
Medical supplies	Ethicon Sutures (AECL)	Italy (1967)	^{60}Co	Unknown
Medical supplies, instruments	Helinos AB of Oskarshawn (H. S. Marsh, Ltd.)	Thoern, Sweden (1967)	^{60}Co	1,000,000 (200,000)
Medical supplies	Becton, Dickinson, and Company	New Jersey		Accelerated electrons
Medical supplies	Hospital Supply & Development Co.	Pittsburgh, Pa. (Service irradiations from other people)		Accelerated electrons

*Unpublished information supplied by John Maddox of USAEC–DID.

Table 2 CHEMICAL PRODUCTION PROCESSES USING RADIATION

Process or reaction	Location	Stage of development	Radiation source	Ref.
Ethyl bromide manufacture	Dow Chemical Co., Midland, Mich.	4 years commercial	3132 curies ^{60}Co (~2506 curies, Oct. 1966)	14, 15
Sulfoxidation of hydrocarbons	Esso Research, Linden, N. J.	Pilot plant	~12,000 curies ^{60}Co	16
Sulfoxidation of hydrocarbons	Farbwerke Hoechst, Frankfurt, West Germany	Pilot plant	~20,000 curies ^{60}Co	17
Oxidation of paraffins	Rumania	Industrial scale	γ rays	18, 19

of hydrocarbons is exposed to the gamma radiation prior to the introduction of the oxygen.

Polymerization Processes

One of the most active areas of radiation processing has consistently been that of initiation of polymerization. A summary of processes known to be either commercial or in intensive development is presented in Table 3. The radiation-induced polymerization of ethylene, while not yet commercial, has received increasing attention. The development work in progress at Brookhaven National Laboratory (BNL) is at the stage where a continuous-flow system has been successfully operated. Work at the Takasaki Radiation Chemical Research Establishment (Japan), by contrast, has been centered on the production of the powdered polyethylene obtained at temperatures below the melting point of the polymer. A significant event is provided by one industrial company in the United States, whose name has not been disclosed, who is in the process of pilot-planting a radiation process for polyethylene.

The practical application of solid-state polymerization has received encouragement from the pilot plant built and operated at the Takasaki Establishment. Here, trioxane is polymerized in the crystalline form by use of 3-Mev-electron irradiation.

A recent burst of activity has been seen in application of radiation to the curing of a variety of polyester-monomer coatings. As mentioned above, two manufacturers have designed accelerators especially for such purposes. Systems for rapid curing of both coatings and laminated forms are now commercially available, and the near future should tell whether actual end-use products will be made routinely by such methods.

The use of radiation to bring about polymerization of various monomers within the structure of wood has formed the center of much discussion and some

Table 3 POLYMERIZATION PROCESSES USING RADIATION

Monomer	Location	Stage of development	Radiation source	Ref.
Ethylene	Brookhaven National Laboratory, Upton, N. Y.	100-ml static, 89-ml flow system	^{60}Co	20
Ethylene	Takasaki Radiation Chemical Research Establishment, Japan	10 liters	^{60}Co	21
Ethylene	USSR	2 liters		22
Ethylene	USA	Pilot plant	^{60}Co	3
Ethylene vinyl compounds	Farbwerke Hoechst, Frankfurt, West Germany		11,000 curies ^{60}Co	16
Trioxane	Takasaki Radiation Chemical Research Establishment, Japan	Pilot plant	3-Mev electrons	21
Polyesters + styrene, acrylics, as coatings	Radiation Dynamics, Inc., Westbury, N. Y.	Commercially available	0.3-Mev electrons	7
Polyesters + styrene, acrylics, as coatings	Ford Motor Company and Boise Cascade Corp., Boise, Idaho	Commercially available	0.3-Mev electrons	7, 23
Polyester resins in glass-fiber laminates	Radiation Dynamics, Inc., Westbury, N. Y.	Available	1.5-Mev electrons	24
Wood–plastic combinations	West Virginia University, Morgantown	Large-scale testing of samples	^{60}Co	25
Wood–plastic combinations	American Novawood Corp., Lynchburg, Va.	10 products ready for market	^{60}Co	10
Wood–plastic combinations	Lockheed-Georgia, Dawsonville, Ga.	250,000 bd ft/year	Reactor; 250,000 curies ^{60}Co	10
Wood–plastic combinations and chemicals	Eurisotop, Euratom (mobile, truck-mounted)	Authorized	~175,000 curies ^{137}Cs	

controversy in recent years.[26] Whether or not conventional catalysts can be used and whether straight impregnation or swelling of the wood structure is the most efficient way to bring about the desired properties, this field can now be said to be entering the commercialization stage. The American Novawood Corp. claims 10 different products are ready for the marketplace, e.g., parquet flooring, tiling, and molding. Lockheed-Georgia is able to process annually by radiation a quarter million board feet of lumber impregnated with monomer.

Graft Copolymerization

Table 4 presents the current status in the field of radiation graft copolymerization. One of the most significant new items is the advanced stage of work on the presumed grafting of monomers to various fibers. Two separate groups, Deering-Milliken, Inc., and a combination of Radiation Research Corp. with Burlington Industries, have announced commercial processes to impart special properties to cloth using radiation. Such processes allow stains to be easily washed out of materials, and the property has been termed "soil release." A more durable press can also be attained by the treatment. Accelerated electrons are the obvious choice for processing these materials. Another application of grafting to fiber materials is illustrated by pilot-plant experiments carried out at the Takasaki Radiation Chemical Research Establishment in cooperation with the Toyo Spinning Company of Japan. Improvement in dyeability and wash-and-wear qualities are imparted by the grafting of styrene or acrylic monomers to the regenerated cellulose fibers.

Polymer Treatments

The direct alteration of polymers by means of high-energy radiation (Table 5) must be considered the earliest truly commercial application of radiation to chemical systems. Irradiated polyethylene tape has been available for more than a decade from the General Electric Company. Cryovac L, a major food-wrapping material, is produced by irradiation of polyethylene film during processing. The properties of such insulating materials as polyethylene are enhanced by a number of companies who irradiate wire, cables, and similar forms on a routine commercial basis. Other fabricated forms of polyethylene

Table 4 GRAFT-COPOLYMERIZATION PROCESSES USING RADIATION

System	Location	Stage of development	Radiation source	Ref.
Polyethylene + acrylic acid	Dow Chemical Co., Midland, Mich.	Pilot plant	2-Mev electrons	27, 28
Polyvinyl chloride + styrene	Dow Chemical Co., Midland, Mich.	Sampling	^{60}Co	29
Polyester–cotton fiber blends + monomers (presumed)	Deering-Milliken, Inc., Blacksburg, S. C.	Commercial, full-scale	Accelerated electrons	30
Polyester–cotton fiber blends + monomers (presumed)	Radiation Research Corp., Westbury, N. Y.; Burlington Industries, Greensboro, N. C.	Commercial	High-voltage discharge	31
Cellulosic fibers + styrene or acrylics	Takasaki Radiation Chemical Research Establishment, Japan	Pilot plant	2-Mev electrons	21
Polyethylene films + vinyl monomers	Sekisui Chemical, Japan	Pilot plant	Accelerated electrons	32

Table 5 POLYMER IRRADIATION PROCESSES

Polymer	Location	Stage of development	Radiation source	Ref.
Natural rubber latex	Saclay, France	1 ton/hr	Accelerated electrons	33, 34
Polyethylene for foam	Toyo Rayon, Japan	Commercial		35
Polyethylene for foam	Sekisui Chemical, Japan	Commercial		
Polyethylene tape	General Electric Company, Pittsfield, Mass.	Commercial for several years	Accelerated electrons	
Polyethylene film	W. R. Grace & Co., Simpsonville, N. C.	Commercial for 7 years	Accelerated electrons	36
Wire and cable coverings, heat-shrinkable plastic tubing	Raychem Corp., (subsidiaries, Sequoia Wire Co. and Rayclad Tubes, Inc.), Redwood City, Calif.	Commercial	Accelerated electrons	11
Polyethylene wire covering and other materials	Electronized Chemicals Corp., Burlington, Mass.; Radiation Dynamics, Inc., Westbury, N. Y.; International Telephone and Telegraph Corporation, Wire & Cable Div., Clinton, Mass. (Formerly, Suprenant)	Commercial	Accelerated electrons	11
Polyethylene wire and cable covering, tubing, film, packing bags	Sumitomo Electric Industries, Japan	Commercial	2-Mev electrons	35
Cable insulation	Germany		Accelerated electrons	37
Polyethylene oxide	Union Carbide	Design of plant	100,000 curies ^{60}Co	10

are commercially produced and irradiated by the Sumitomo Electric Industries of Japan. Tubing, films, and bags are included in this category.

Another application of high-energy radiation to polyethylene lies in the area of foamed sheet. While the exact technique has apparently not been revealed, commercial samples of foamed polyethylene, the processing of which requires radiation, are available from both the Toyo Rayon Company and the Sekisui Chemical Company of Japan. Presumably the cross-linking ability of radiation is useful in the manufacture of these materials.

While polyethylene cross-linking is usually performed on the structural form of the polyethylene, another application of radiation successfully cross-links rubber latex while still in the fluid form. Quantities as large as 1 ton/hr have been produced at the Saclay establishment in France.

Another very recently announced application of radiation to alter polymers involves polyethylene oxide. Union Carbide Corporation is reported[10] to be designing a plant that could use as much as 100,000 curies of ^{60}Co for the purpose of altering the molecular weight of this polymer. If this process continues to the truly commercial stage, it will constitute the largest single use of a radioisotope for chemical or polymer processing.

Other Applications

As the field of technology grows into the realm of commercial reality, the amount of commercial secrecy surrounding operations necessarily increases. The reason is the great importance of obtaining a competitive advantage in a field where large sums of money are to be invested. It is certain that other radiation processes are in stages of intense development and may even be on the threshold of commercial production. Furthermore, facilities are now available for custom irradiation of materials on a confidential basis. Much of the current irradiation of semiconductors is done on this basis, and it is very difficult to determine the true volume of samples being processed.

Calculation Procedures

As the number of processes entering intensive development increases, the requirement for quantitative relationships between sources and target become more important as well. A report[38] on computational methods for various arrangements of sources and absorbers was prepared by personnel at BNL. It is particularly useful in the solution of problems involving planar sources. Furthermore, this program has been tested with actual measurements around a radiation source, so its reliability is quite good. Another recent paper[39] has dealt with the problem of optimizing the geometry of radiation sources within chemical reaction systems. With publications such as these now appearing in the literature, the engineering of radiation facilities for specific purposes can be put on a more nearly exact basis. (MG)

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SINB and USAEC Hold Food-Irradiation Briefing for Industry at Oak Ridge

On Feb. 2–3, 1967, the Southern Interstate Nuclear Board (SINB) and the AEC held a briefing on food irradiation in Oak Ridge, Tenn., for industry. This was the sixth in the series of industrial briefings on atomic power in industry sponsored by SINB. There were 217 registrants and 24 speakers, prominent among whom were James L. Goddard, Commissioner, U. S. Food and Drug Administration (FDA), and Samuel M. Nabrit, Commissioner, U. S. Atomic Energy Commission.

Commissioner Nabrit delivered an overview of food irradiation, and he chided both FDA and the food industry for not actively furthering the commercialization of the process. He pointed out that the food industry spends only one-tenth as much on research and development as the overall industrial average.

The Army program of research on safety of irradiated foods was reviewed by E. S. Josephson, U. S. Army Radiation Laboratory. The AEC program of safety research was covered by L. A. Whitehair, Division of Biology and Medicine, AEC. The views of the FDA on safety were presented by R. S. Roe, Bureau of Science, FDA; the stringency of FDA requirements was underlined. He also revealed that FDA has decided that 90-day feeding studies cannot be substituted for 2-year feeding studies, even when the 2-year feeding studies were done on closely related foods. This had been suggested for some fruits. The decision reflects FDA's growing cautiousness about clearing irradiated foods. S. R. Hoover, Agricultural Service, U. S. Department of Agriculture (USDA), summarized USDA attitudes toward food irradiation.

John T. Conway, Executive Director for the Joint Committee on Atomic Energy, presented the luncheon address. He, like Commissioner Nabrit, was critical of industry's slowness. He made a plea for better communications between FDA and petitioners. After lunch, progress in technology was reported. E. C. Maxie, University of California, discussed radiation treatment of fruit; A. F. Novak, Louisiana State University, covered irradiation of fish; J. H. Powers, University of Georgia, reviewed irradiation of grain and potatoes; E. Wierbicki reported on meat irradiation at the U. S. Army Natick Laboratories; and C. W. Schroeder, Thomas J. Lipton, Inc., described Lipton's patented process of irradiation of dehydrated vegetables used in dried soups.

A keenly anticipated address at the briefing was that of Dr. Goddard. His talk at dinner that evening received considerable attention in the popular press, and it is reproduced *in toto* below. Before the dinner, irradiated bacon wrapped around chicken liver was served as hors d'oeuvres. Baked potatoes irradiated to inhibit sprouting were served with the meal as were biscuits made from irradiated wheat flour. The comments of the diners are summarized in the article following Dr. Goddard's address.

The second day, J. H. Frankfort, Parsons-Jurden Corp., and D. A. Trageser, High Voltage Engineering Corporation, described present facilities for food irradiation. D. Streever, Daniel Yankelovich, Inc., gave a preliminary summary of the report by Daniel Yankelovich, Inc., indicating that consumers will accept irradiated food with little difficulty provided FDA approves it and there is a known brand name on the package. (This report, NYO-3666-1, will be covered fully in the next issue of *Isotopes and Radiation Technology*.) The interest in irradiation of fish shown by commercial purveyors of seafoods was described by J. D. Kaylor, Bureau of Commercial Fisheries. The economics and costs of food irradiation were covered by H. W. Ketchum, U. S. Department of Commerce. W. L. Brown, John Morrell & Co., presented a short description of the irradiation of bacon at Brookhaven National Laboratory's High Intensity Radiation Development Laboratory to fulfill Morrell's contract with the Army. (F. E. McKinney)

Good Science and Good Food

By James L. Goddard, M.D.*

I am delighted to appear on this SINB program and to be in such distinguished and gracious company. Let me congratulate Chairman John Breckinridge and Nat Welch and the many other members of this group who planned and are carrying out this 2-day briefing. I'm sure that everyone who attends will gain yet a deeper understanding of the power of atomic energy and—even more important to every American—the power of peace.

You may recall that President Johnson told the Congress and the people, in his State of the Union message last month, that "we are eager to turn our resources to peace, our efforts in behalf of humanity." I am positive that every person here this evening shares that eagerness with our President. But we know that real progress can only come after careful planning, after responsible research and development, and after solid capitalization have been achieved.

*Commissioner of Food and Drugs.

These activities take time. They must be started and carried on as if peace were imminent—as, indeed, we hope it is. So I presume that I am speaking this evening in the company of Americans who are not only interested in the power of the atom—but also in the power of man's mind and spirit as well.

The use of radiation to preserve food is a comparatively new idea. Granted that the first patent for an ionizing process was issued back in 1930 in France, it was not until World War II that some support for research went into this area. Later, in the 1950's, the Atomic Energy Commission and other agencies of the federal government began to probe more deeply into the usefulness of radiation for food preservation and food surface sterilization. Our own Food and Drug Administration became involved as well, through the work of Drs. Lehman, Laug, Nelson, and others.

With the passage of Public Law 85-929 in September of 1958, the FDA became a central agency in the team of agencies involved with food irradiation. That law is the so-called "Food Additives Amendment" to the Federal Food, Drug, and Cosmetic Act. It is not unknown to anyone in the audience, I am sure; so I will not dwell upon it.

I am, however, reminded of several statements published back in 1965 by the Joint FAO/International Atomic Energy Agency/WHO Expert Committee.[1] You are all familiar with the Technical Report, I am sure. It says that "Before any legislation is enacted to permit irradiation of food, there should be clear evidence that any disadvantages that might possibly arise from the use of the process are substantially outweighed by the expected advantages."

By then the United States already had on the books the Food Additives Amendment, which was enacted in the spirit of that statement, even though the Joint Committee's work came several years later. This particular paragraph also contains the following sentence: "In particular, no known hazard to health"—and I would like to underscore that—"no known hazard to health should be introduced either during application of the treatment or in the utilization of the product."

Surely this is extremely cautionary language and cannot be read with inattention. We believe the present United States law must be carried out in the spirit embodied in these sentences. Our regulations follow this spirit. I intend that they should continue to do so.

Let me move on to a few additional points raised by the Joint Report. I am particularly drawn to the section on testing. It is an eminently reasonable and progressive section, signifying caution to the investigator but in no way stifling to creative experimentation. It says, for example, that animal experimentation must be carried out, however limited it might be. Man—as we all know—is the ultimate subject. "Nevertheless," says the report, "the absence of adverse effect in animals fed irradiated food can be used as a reasonable basis for predicting the safety of the process when applied to food intended for human consumption." The report continues—and this will be a final quote for a while—"The tests which must be applied to an irradiated food to establish its safety for consumption are broadly similar to those generally applied to ensure the safety of food additives."

What do these random sentences add up to? Why should they catch our attention? I think they are worth noting because they define the parameters, as it were, of the problem of getting food-irradiation petitions past the FDA. I could state the ideas one way or another; I chose, however, to use the FAO/IAEA/WHO language, for many of your colleagues and some here in this audience this evening participated in that Joint Expert Committee meeting in Rome, from which the language was drawn.

First, then, if we are to have regulations under the irradiation law, they must be ones which prohibit any "known hazard to health" from being introduced into our food supply. Second, if we are to test in this area, we must seek the "absence of adverse effect in animals," in order to proceed with any assurance to man.

The burden of proof, then, is indeed heavy, and I can understand that it would be discouraging as well to many a researcher. This leads me to the conclusion that beyond the need for solid, well-founded research results is a need for solid and well-founded support for good research personnel as well.

The food-irradiation field is young and, like any young science, can suffer growing pains that are hard to dismiss. There may be a tendency among some of us to be impatient with data, with animal populations, with our own colleagues, and with others in the field as well—in private or in governmental agencies.

We should recognize that this can happen, and we should not be overwhelmed by it. Those of us who bear the final responsibility for either submitting research and completed petitions or who have the final responsibility to pass upon those petitions must not let the normal growth processes of young science produce unfounded prejudices in our judgment. Neither can we lean the other way and relax the safeguards carefully built for the protection of the public in order to give this particular field an additional—and an unjustifiable—margin of possible error.

Basically I think we are moving into an area of great promise. But this audience of experts, I am sure, knows better than I that this is also an area of potential hazard. How do we balance out the promise against the elimination of the hazard? That very question seems to be the fundamental one at this moment, and we must turn to it together.

I am pleased to say that Chairman Seaborg of the Atomic Energy Commission and I have already dis-

cussed this matter. We both agree that more discussion is certainly needed, not only between the two of us but at our staff levels as well. We both realize that the FDA has exercised great caution in approving irradiated-food petitions. Dr. Seaborg and I also understand the many factors — scientific, professional, and personal — that sometimes enter the research and petition process.

These are natural, but they should be carefully weighed. At some point we have to decide, together, that this new field will mature under the best scientific guidance possible and with the assistance — not the obstruction — of both the Atomic Energy Commission and the Food and Drug Administration. We have to bring to light the forward-looking research; we have to identify the real scientific leaders in this field; we have to build better communications throughout this field so that prejudice and a lack of balance will not prevail, but that good judgment will.

I am making this point so strongly this evening because I saw from the list of distinguished guests that I had, in fact, a captive audience. I would like very much to capture you and start you reflecting upon the work that has been carried on already in this field and about the work that lies ahead.

We have learned a great deal from the work of the last half-dozen years or so. And no one would argue that it has all been perfect. But certainly enough data have been developed to give us grounds for hope. There have been a number of new starts that are yielding even more valuable data. The X radiation of wheat, the electron-beam process for inhibiting sprouts on white potatoes, the new work in packaging — all these new developments are keeping up the momentum.

At the level of the researcher, there are a number of things we should try to accomplish. First, I think we must constantly reemphasize the terms of the law and the regulations that bind us. I can tell you that this is one of the most important areas of our work in the *drug* field: impressing upon researchers and clinical investigators that our agency and the law itself are not the results of caprice. It is all a very serious business. And we evaluate the data of all researchers with great care. There is delay, yes. There are differences of opinion, yes. But there will not be obstruction or prejudice in the handling of the data at our end. I am not aware that there has been and there will *not* be.

Second, it would seem that good communications in this field might be given some serious new consideration. It can be very discouraging for industry executives and the consumer as well to be led in any way into thinking that this or that process for this or that commodity is in effect today, when this may not be the case. We have to be fair-dealing with each other and with the public. Communications among researchers, among the government agencies involved, among business and industry, and between all these groups and the consumer should be carefully presented so that expectations do not exceed capabilities. Setbacks may come at any time. Why, then, should we do anything to build in discouragement and disillusion beforehand? Clearly, we should not.

Third, and this would be my last general thought at this time, I would encourage everyone to reflect once again on the role our nation plays in food-processing leadership. The growers and packers and distributors of foodstuffs the world over watch what is accomplished here in the United States. Our laws are taken to be model laws. Our business practices are imitated — sometimes with extremely good effect for others and pressing competition for our own firms.

If "imitation is the highest form of flattery," then this nation has been flattered almost to excess by those in the food field in other lands. As the ability to convert atomic energy into peaceful uses spreads among more countries, our experiences with irradiated-food processes will be not only carefully watched — as they already are — they will be carefully copied, as well. A generation has passed — a quarter of a century — since world leadership was thrust upon the United States. Most Americans may tend to think of this leadership in very large-scale terms: winning a war, shooting for the moon, stopping Communist expansion. But leadership comes in details as well, such as setting international standards for food additives. In this matter, our government is a strong participant in the Codex Alimentarius[2,3] effort, although the American people may never become generally aware of that. But we are, and in this matter of irradiated foods we have the opportunity to make a significant and lasting contribution. We can do that, if we maintain high standards in our own domestic activity, in research, in administration, and in industrial processing as well.

The standards our agency has set for business and industry are high, but they are not unreasonable, as I have noted earlier. In fact, we have very often adopted standards and guidelines that industry has — in conjunction with science — set for itself. Certainly the FAO/IAEA/WHO Technical Report is a case in point. There are others in both the food and drug fields to which I could point. So I would ask this audience to consider that the standards of food-irradiation research and the conditions under which irradiation processes may be approved have to be of the highest caliber. "Disadvantages" must be "substantially outweighed by the expected advantages." There should be "the absence of adverse effect in animals" before moving on to men.

I would like to close with one final observation. As you know, the old Metropolitan Opera House in New York City is being torn down, to the regret of many. Not long ago a friend dropped by my office with a red-and-gold-covered souvenir menu from Louis Sherry's Metropolitan Opera Restaurant. Sherry's

was, of course, as much an institution as the Met itself. The menu was not especially exotic—lamb chops, filet of Boston sole, breast of chicken and Virginia ham, spinach, lima beans, and so on. Occasionally you came upon veal Renata and mushrooms Bercy, and, naturally, cerises flambé. But in the main, this menu seemed to be straightforward, reliable, and quite dignified. Friends who had dined at Sherry's have spoken of its fresh-cooked, wholesome food and its good sauces.

As I say, Louis Sherry's is gone. The Met itself is coming down. In the new Lincoln Center Opera House a restaurant called the "Grand Tier" is now operating, run by the United Canteen Corporation. I hope one day to sample its fare since I never made it to Louis Sherry's.

But I wish to observe that those who are in the food field must remain aware of what they do when they stimulate change. Change for the sake of change is not good enough. If we give up Sherry's, we expect something better. If we are to choose between irradiated foods and, for example, nonirradiated frozen or canned foods, we will as consumers as well as regulators, choose what is better—better scientifically and, if possible, aesthetically as well.

The burden of proof is heavy—but it does weigh upon the innovator. Those who are deeply involved in food irradiation must answer the hard questions of vitamin and mineral preservation, of the effect upon harmful and useful microorganisms in food, of teratogenicity among animals—including man—and of sterilization of the reproductive process itself. The answers must be good answers—the best that can be obtained. And they must be obtained in the true spirit of discovery—not to prove an advance prejudice.

The peaceful uses of the atom are manifest. We, with our limited vision clouded by several wars, cannot really see all the promise of this energy source. But we can pave the way now—through good science—for our younger colleagues to take up the challenge and work *with* us and *after* us on expanding our food supply through radiation processes. Together, let us do the job right.

Thank you. (FEM)

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Summary of Comments Made on Irradiated Food Served at SINB Briefing

All three irradiated foods served at the Southern Interstate Nuclear Board briefing at Oak Ridge, February 2, received high preference ratings at the dinner attended by almost 250 people. The bacon had been irradiated with 4.5 to 5.6 Mrads between June 27 and July 8, 1966, and stored without refrigeration since then. The potatoes had received 10,000 rads, and the wheat flour had been given 40,000 rads at the U.S. Army Natick Laboratories. A 9-point hedonic scale was used on the cards passed out at the meal, with 9 meaning "Like extremely," 1 meaning "Dislike extremely," and 5 meaning "Neither like nor dislike."

The bacon was served in two ways. First, it was served wrapped around chicken livers as hors d'oeuvres at the social hour. Some attendees objected to the bacon taste being disguised by the liver taste. E.S. Josephson produced a package of uncooked bacon, and after the hors d'oeuvres were eaten, several people tried to fry the bacon in the chafing dish—with little success. The bacon was also served as a garnish on a club steak at the dinner; the bacon was crisper than when served with the chicken livers. The bacon received a 7.43 average rating, based on 214 cards returned after the meal; this ranks it between "Like moderately" and "Like very much." Fifty of the cards had comments on them. These ranged from "All the way with the FDA" to "Has good 'liver' flavor" to "Typical G.I. bacon." Many of the diners thought the bacon should be crisper; only two people thought they could definitely distinguish an "irradiation taste."

The potatoes were baked and served in aluminum foil. They received an average rating of 7.31; 215 cards were returned. The 35 comments objected variously to cooking the potatoes in foil, to eating such a high-calorie food, and to the potatoes' being both overcooked and undercooked. No irradiation taste was noted.

The hot rolls made from irradiated flour received a lower preference rating than either the bacon or the potatoes. Several of the comments suggested that preparation in the kitchen might have been responsible. The 6.79 rating was based on 200 responses. The 46 comments generally disagreed with the menu as to the form in which the flour was served—"In Georgia these are biscuits"—and objected to the fact that they were burned on the bottom and underdone. Several kinds of off-tastes were noted, but one commenter said, "Better than I got during World War II."

(F. E. McKinney)

Section IV

Isotopes and Radiation Technology

Life Sciences

Iodine-123: Development and Usefulness of a New Radiopharmaceutical

By Buck A. Rhodes,* Henry N. Wagner, Jr.,* and Martha Gerrardt

Abstract: The properties of ^{123}I, its production, and uses are reviewed.

In the past, the radionuclides used in biology and medicine have been chosen primarily because they were available rather than because they were most suitable for the problem at hand. However, since it is now possible to produce a much wider selection of nuclides, it is becoming feasible to select the best nuclide for a given medical application.[1-4] The steps in making such a selection are (1) specification of the optimum chemical and physical properties that a radionuclide should have to most effectively solve the problem at hand; (2) choice of the radionuclides approximating the ideal from the list of about 1500 known ones and arrangement of them according to relative values of the information to be obtained with them and the associated radiation exposure to the patient; and (3) estimation of the production costs of the radiopharmaceutical. On the basis of these considerations, the most suitable radiopharmaceutical can be chosen.

A radionuclide is valuable for the in vivo counting needed in medical work if it has a high emission rate of photons of proper energy for collimation and efficient detection and if it concentrates preferentially in or surrounding the tissue of interest. It also must give a low radiation exposure to the patient, i.e., have as short a half-life as practicable for the test and give a low-energy deposition rate in tissue.

In this report we summarize the evidence that indicates ^{123}I to be superior to other isotopes of iodine for many biomedical purposes.[1,3-6] Although this isotope has not previously been generally available, its commercial production is now possible,[7] and we can anticipate that it will play an important role in medicine.

Properties of ^{123}I

Iodine-123 decays[5,8,9] by electron capture, yielding ninety-two 28-kev X rays and eighty-four 159-kev gamma rays per 100 disintegrations (Fig. 1). It emits more photons per disintegration than ^{131}I, and a higher fraction of these are in the energy range suitable for detection with internal scintillation detectors. Since the chemical behavior of all iodine isotopes is

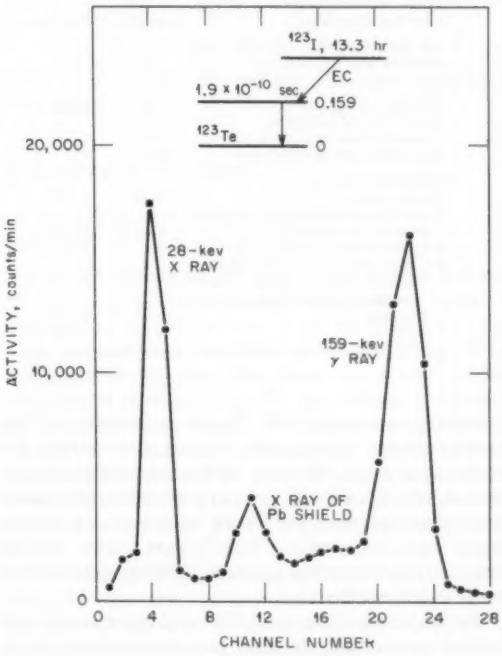

Fig. 1 Decay and photon spectrum of ^{123}I. The spectrum was obtained with a 512-channel gamma-ray spectrometer, using a 5- by 5-in. NaI(Tl) crystal located about 4 in. below the ^{123}I source.

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the same, we can select the radioisotope to be used on the basis of physical data alone. For example, when iodine is used in thyroid tests and its activity must be measured 24 hr after administration to the patient, the optimum physical half-life for such a test can be shown to be 16.6 hr. Since ^{123}I has a physical half-life[10] of 13.30 ± 0.05 hr, it has the most nearly ideal half-life of any isotope for tests in which the isotope is administered one day and the activity is counted the next.

Iodine-123 releases an average of 27 kev energy[2,5,9] per disintegration which is absorbed within a few microns of the nucleus. Of the other isotopes of iodine, only ^{125}I releases less absorbed energy per disintegration—an average of 24 kev. Iodine-131, on the other hand, releases seven times as much absorbed energy. The low rate of energy deposition in tissue, together with a nearly optimum physical half-life, makes ^{123}I an attractive iodine isotope for minimizing radiation exposure to patients during clinical use.[11] In Table 1 dosimetry data for ^{123}I are compared with those of ^{131}I.

Table 1 DOSIMETRY DATA FOR ^{123}I COMPARED TO ^{131}I

	^{123}I	^{131}I
Half-life, days	0.5545	8.08
Average beta plus beta-like energy, Mev/disintegration	0.027	0.187
Point-source gamma-ray dose-rate constant $(cm^2)(r)/(mc)(hr)$	1.5	2.18
X ray		
Average energy, Mev	0.028	0.029
Probability of emission, photons/disintegration	0.92	0.02
Half-thickness in water, cm	1.7	1.7
Principal gamma ray		
Energy, Mev	0.159	0.364
Probability of emission, photons/disintegration	0.84	0.80
Half-thickness in water, cm	4.7	6.3
Intrinsic efficiency of detection 3 cm from a 3- by 3-in. NaI crystal, pulses/photon		
Photopeak	0.92	0.53
Total	0.92	0.735

Another advantage of ^{123}I is suitability of the emitted photon energy for making corrections for tissue absorption. Because of the low photon energy, the emitted X ray will be more strongly attenuated by tissues overlying the target than will the higher energy gamma photon. Thus target depth can be obtained by comparing relative counting rates of the X and gamma activities.

The major problem with ^{123}I at present is its high cost of production. It also has the problem of all short-lived isotopes: more frequent shipments, standardizations, and dose preparations make its use costly. However, basic production costs may be reduced if production becomes more widespread and the techniques of handling short-lived isotopes in the clinic are simplified as they are used more and more. The advantages of ^{123}I may be summarized as follows:

Physical:

1. Low radiation dose to patient due to (*a*) short half-life, (*b*) absence of particulate emissions during decay, and (*c*) low dose-rate constant.
2. High yield of detectable photons due to (*a*) high abundance of both X and gamma rays, (*b*) low tissue absorption of gamma rays, and (*c*) high efficiency of detection of X and gamma rays.
3. Simplified collimation and counting due to (*a*) convenient energies of the X and gamma rays and (*b*) simple or "clean" gamma-ray spectrum.

Chemical:

1. Tracer of biologically important element.
2. Versatile tagging agent.
3. Tagging methodology already worked out with other iodine isotopes.
4. Preparations having very high specific activity can be made.
5. Carrier-free preparations are possible.

Clinical:

1. Short half-life allows for (*a*) repetitive testing and (*b*) low interference with other radioisotope tests.
2. Target depth can be estimated by measuring both the X- and gamma-ray counts.
3. Better counting statistics and/or reduced patient exposure.
4. Automatic, rapid cleanup of any spills.

Production of ^{123}I

Iodine-123 made its first appearance in the Table of Isotopes[12] in 1950 following the discovery[13,14] of an activity with a 13-hr physical half-life in the product of an $(\alpha, 2n)$ reaction on ^{121}Sb.

Iodine-123 has been produced[7,15] by the Isotopes Development Center at Oak Ridge National Laboratory (ORNL) by bombarding ^{123}Te with 15-Mev protons. Tellurium that had been enriched by an electromagnetic process to approximately 80% ^{123}Te was obtained for target material at an initial cost of about $3500 for 270 mg. The low natural abundance of ^{123}Te, 0.87%, contributes to the high cost of target material, but fortunately the unconverted ^{123}Te is reusable. Yields of 80 to 100 mc of ^{123}I per cyclotron beam-hour are obtained, and about 70% of the ^{123}I is recovered from the target. Allowing two half-lives for processing and shipping, approximately 15 mc of usable ^{123}I is produced per cyclotron beam-hour. Production runs of up to 5 hr appear to be feasible. The Isotopes Development Center has received approval from the USAEC to produce and distribute ^{123}I. The product, which will be produced

in the 86-in. cyclotron, will have the following properties:

Half-life	13.02 hr
Production method	$^{123}Te(p,n)^{123}I$
Chemical form	NaI in ~0.5*M* NaOH
Concentration	~1 mc/ml at time of processing (other concentrations may be available on request)
Specific activity	No added carrier
Purity	>99% exclusive of other iodine activities (^{124}I, ^{125}I, ^{126}I) (^{124}I concentration supplied with each shipment)
Other iodine activities	<3% of total radioactivity at time of processing

The price will range from $15 to $25 per millicurie. Details may be obtained from Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37830.

Iodine-123 has also been produced at the Brookhaven National Laboratory by bombarding ^{123}Sb with 30-Mev ^{3}He particles. The yield was 173 $\mu c/\mu a$-hr, or approximately 5 mc per cyclotron beam-hour. The cross section for the $^{123}Sb(^{3}He,3n)^{123}I$ reaction, 800 mb, is 100 times as great as that of the competing reaction, $(^{3}He,2n)$, which gives rise[16] to the ^{124}I impurity. A natural-antimony target, which is 42.75% ^{123}Sb, was used in test runs. However, antimony enriched to 95–99% ^{123}Sb is available at $2.50 per milligram.

Iodine-123 has also been reported[17–19] in the product of the reaction of 600-Mev protons with cesium, neodymium, or samarium; of 590-Mev and 18-Gev protons with lanthanum or uranium; of 18-Gev protons with tantalum or gold; and of 100- to 660-Mev protons with ^{127}I.

Reference to a chart of the nuclides reveals that there are at least 20 hypothetical ways of directly converting stable neighbors—xenon, tellurium, and antimony—to ^{123}I:

$^{124}_{54}Xe(\gamma,p)^{123}_{53}I$	$^{122}_{52}Te(p,\gamma)^{123}_{53}I$
$^{124}_{54}Xe(n,pn)^{123}_{53}I$	$^{122}_{52}Te(^{3}He,dn)^{123}_{53}I$
$^{124}_{54}Xe(t,\alpha)^{123}_{53}I$	$^{122}_{52}Te(t,3n)^{123}_{53}I$
$^{126}_{54}Xe(\gamma,t)^{123}_{53}I$	$^{122}_{52}Te(d,2n)^{123}_{53}I$
$^{126}_{54}Xe(p,\alpha)^{123}_{53}I$	$^{123}_{52}Te(p,n)^{123}_{53}I$ (used at ORNL)
$^{120}_{52}Te(\alpha,p)^{123}_{53}I$	$^{124}_{52}Te(d,3n)^{123}_{53}I$
$^{120}_{52}Te(t,\gamma)^{123}_{53}I$	$^{124}_{52}Te(p,2n)^{123}_{53}I$
$^{122}_{52}Te(\alpha,t)^{123}_{53}I$	$^{121}_{51}Sb(\alpha,2n)^{123}_{53}I$ (Refs. 13,14)
$^{122}_{52}Te(t,2n)^{123}_{53}I$	$^{121}_{51}Sb(^{3}He,n)^{123}_{53}I$
$^{122}_{52}Te(d,n)^{123}_{53}I$	$^{123}_{51}Sb(^{3}He,3n)^{123}_{53}I$ (used at BNL)

The literature survey suggests that many of these reactions have not yet been investigated. Indirect methods of preparing ^{123}I could also be explored (Fig. 2). If ^{123}Cs or ^{123}Xe should be synthesized, ^{123}I would grow in as the parent ^{123}Xe or grandparent ^{123}Cs decays.

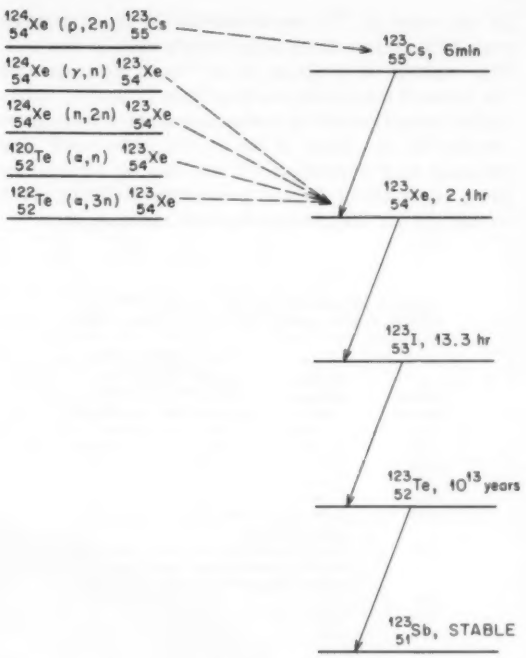

Fig. 2 Indirect methods of synthesizing ^{123}I, postulated from known types of nuclear reactions and the decay series of which ^{123}I is a part.

Examples of ^{123}I Uses

MEASUREMENT OF THYROID STRUCTURE AND FUNCTION

The superiority of ^{123}I over other iodine isotopes for the clinical measurement of the 24-hr uptake of radioiodine by the thyroid is shown by the calculations summarized in Table 2. If sufficient ^{123}I is administered to give the same count rate as that obtained at present with ^{131}I, the radiation exposure is decreased by a factor of approximately 35. If the uptake of radioiodine is measured after a shorter interval, the advantage over ^{131}I is even greater. Even when ^{123}I contains certain isotopic impurities, it remains superior to ^{131}I for several days (Figs. 3 and 4).

Not only is ^{123}I safer than ^{131}I, but it also makes possible increased precision in the measurements of the thyroid uptake of radioiodine. A major reason for the loss of precision is patient-to-patient variation in the depth of the thyroid gland in the neck. The magnitude of this error for various iodine nuclides is illustrated in Table 3, from which it can be seen that the lower the photon energy, the greater the error.

In the case of ^{125}I, the magnitude of this error has kept this nuclide from being widely used in this test. The studies of Wellman et al.[20] have demonstrated the value of increasing the precision of thyroid iodine uptake measurement by measuring and correcting the results for the depth of the thyroid in the neck. The increase in precision in ^{123}I use can be obtained by using the ratio of X- and gamma-ray counting rates to correct for the average thyroid tissue depth.

Table 2 RELATIVE MERIT OF ISOTOPES OF IODINE FOR A 24-HR THYROID UPTAKE TEST*

Iodine isotope	Photon energy,† Mev	Count, relative‡ No. of detectable photons/(min)(rad)	
		Differential	Integral
123	0.159	35.1	42.6
124	0.511	0.6	1.8
125	0.028	3.2	--
126	0.386	0.4	1.0
128	Half-life too short for 24-hr uptake measurement		
129	Specific activity too low for accurate 24-hr uptake measurement with ordinary external counting equipment		
130	0.54	2.1	6.2
131	0.364	1.0	1.0
132	0.67	0.03	0.08
133	0.53	11.6	1.5

*Data calculated by Buddemeyer and Rhodes, The Johns Hopkins Medical Institutions, Baltimore, 1966.

†Where more than one photon is emitted, the one giving the highest number of detectable photons per disintegration was used.

‡Relative to ^{131}I. "Detectable photons" are those which escape the thyroid and its overlying tissue and which would be detected by a 3- by 3-in. NaI crystal 10 cm from the source. For differential counting, photons escaping without attenuation were assumed to be detected with the intrinsic photopeak efficiency of the crystal; for integral counting, any photons not fully absorbed were assumed to be detected with the total intrinsic efficiency of the crystal. The value was computed for measurements made 24 hr after administration of the isotope. The dose in rads is the cumulative thyroid self-dose, including that during the period of biological uptake and continuing until biological and/or physical decay was complete.

Myers[21] has obtained both X- and gamma-ray scans of the thyroid with ^{123}I. In scanning, the photon yield and energy are of even greater value than in functional evaluation alone since resolution and contrast of present images are limited primarily by a shortage of emitted photons.

The use of ^{123}I in place of ^{131}I at present prices would increase the costs of thyroid-function tests. Iodine-131 now costs $0.08 per test, and ^{123}I would cost $5 to $6. If the Johns Hopkins Radioisotope Clinic should receive a weekly shipment of ^{123}I at its present cost, the use of this nuclide instead of ^{131}I would increase the overall cost per test 18 to 55%. However, a greater demand for ^{123}I could well decrease its cost.

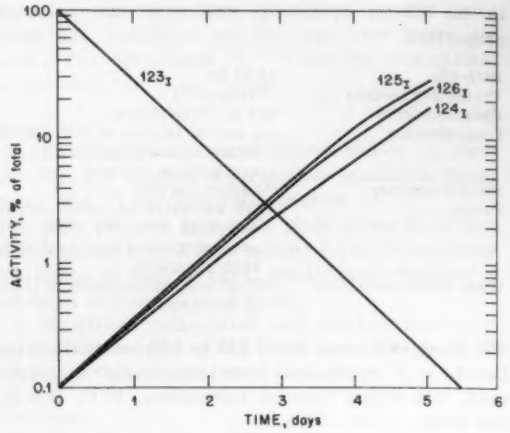

Fig. 3 Iodine-123 decay curve. Also shown are curves giving the percentage contamination of a ^{123}I source that contains an initial contamination of 0.1% ^{124}I, ^{125}I, or ^{126}I. As the ^{123}I source decays, the percentage of radionuclide contamination increases because these contaminants have longer half-lives than ^{123}I.

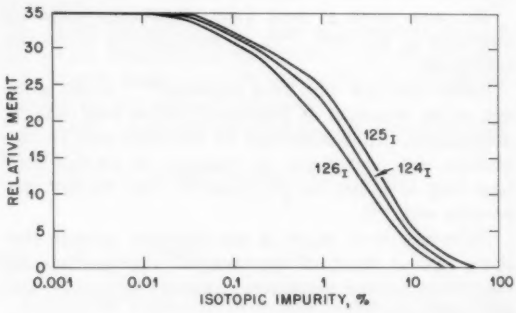

Fig. 4 Curves showing relative merit of ^{123}I containing a single longer lived radioiodine impurity compared to pure ^{131}I for use in the 24-hr measurement of thyroid uptake of radioiodine. If a ^{123}I source contains one or more of these impurities (^{124}I, ^{125}I, or ^{126}I), its relative merit decreases as the percentage of the total activity due to the impurity increases. When (a) the ^{124}I impurity reaches 37% of the total activity, (b) the ^{125}I reaches 53%, or (c) the ^{126}I reaches 30%, the relative merit of the ^{123}I is no longer greater than that of ^{131}I for this test. These percentages apply at the time the radiopharmaceutical is administered to the patient.

BRAIN SCANNING

Matthews[1] has suggested that ^{123}I-labeled albumin may be the best of currently known agents for brain scanning. One potential advantage of it over pertechnetate-^{99m}Tc, an agent widely used for brain scanning, is its longer physical half-life (13.3 vs. 6 hr). This makes scanning possible 24 hr after administration of the dose. Figure 5 shows the first brain scan made using ^{123}I-albumin.

Table 3 ESTIMATED ERROR OF RADIOIODINE UPTAKE MEASUREMENTS DUE TO VARIATION OF THYROID DEPTH

Estimated thyroid depth,* cm	>3.5	3.5	3.0	2.6	2.1	1.8	1.5	1.2	<1.2
Percentage of patients having this depth†	1.9	3.8	9.4	32.1	17.0	13.2	11.3	7.2	3.8

Isotope	Photon energy, kev	Expected % deviation from standard‡						
^{125}I	28	−40.9	−21.3	0	+33.3	+51.1	+86.7	+126.7
^{123}I	159	−20.8	−10.4	0	+16.7	+25.0	+35.4	+47.9
^{131}I	364	−19.9	−8.8	0	+12.1	+21.0	+30.4	+40.0

*Distance between the surface of the neck and the apparent center of activity of each thyroid lobe.

†This distribution of apparent thyroid depths within the neck is that found in the patients studied by Wellman et al.[20]

‡In a study of 53 patients, Wellman et al.,[20] Nuclear Medicine Group, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, found the mean distance between the neck surface and the center of activity to be 2.6 cm. This value was taken as the standard, and the expected % error is relative to it. The error values were determined from efficiency-vs.-distance curves for thyroid counting. K. Foelber, The Johns Hopkins Medical Institutions, 1966, prepared the efficiency curves for these iodine isotopes.

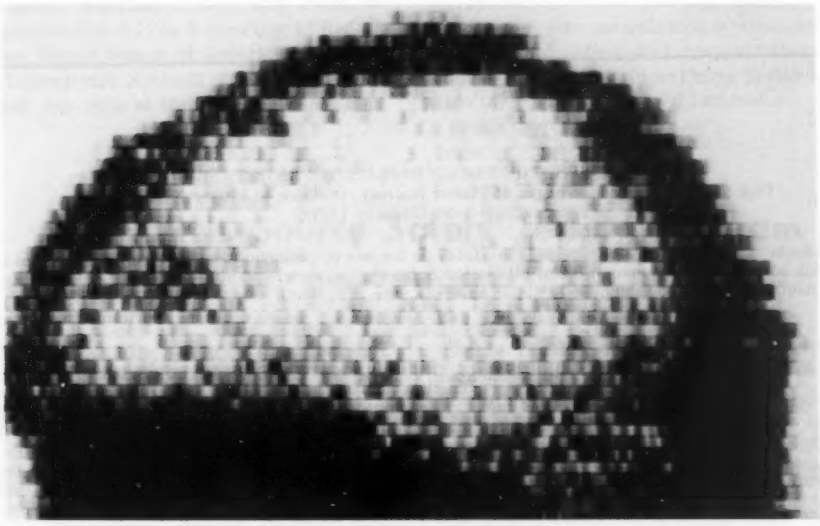

Fig. 5 Brain scan made using ^{123}I, which delineates a tumor in the frontal lobe. This was the first such scan to be obtained with ^{123}I. The ^{123}I was prepared by the ORNL Isotopes Development Group. The radiopharmaceutical, ^{123}I-labeled human serum albumin, was prepared by H. S. Stern of the Department of Nuclear Medicine, The Johns Hopkins Medical Institutions, 1966.

THERAPY

Although evaluation of ^{123}I for therapy has been reported,[22] the results were not stated. This application does not seem to be promising because of the low rate of in situ energy deposition.

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Abstracts of some 200 references (1963–1966) on the use of radionuclides in therapy as internal radiation sources — solutions, colloids, oils, microspheres, and wire implants — are arranged by subject. The nuclides discussed include ^{131}I, ^{32}P, ^{198}Au, ^{182}Ta, ^{90}Sr, ^{90}Y, ^{103}Pd, ^{192}Ir, ^{60}Co, ^{137}Cs, and ^{169}Yb.

(Martha Gerrard)

RADIOISOTOPE TECHNIQUES IN THE STUDY OF PROTEIN METABOLISM

The status of radioactive tracers for the study of protein metabolism was the subject of an International Atomic Energy Agency (IAEA) panel meeting* attended by 13 invited experts from 10 countries and 3 representatives of the World Health Organization (WHO) and held in Vienna, Austria, June 1–5, 1964. At each of the four panel sessions, working papers were presented and discussed in detail. Both the working papers and extensive summaries of the discussions are presented in the published findings.

At the first session, Methods of Preparation of Labelled Proteins and Protein-Like Substances, papers were presented on the preparation of ^{131}I-labeled hormones, proteins, plasma proteins, and polyvinylpyrrolidone; ^{125}I-labeled plasma proteins; ^{51}Cr-labeled albumin; and ^{3}H-labeled proteins. At the second session, Radioisotope Techniques for the Study of Protein Metabolism, there were papers on protein turnover studies, on the measurement of the synthesis rate of liver-synthesized proteins in animals, and on the use of ^{15}N for the study of protein metabolism. At the third session, Radioisotope Techniques for the Study of Gastrointestinal Protein Absorption and Loss, the papers included the study of protein absorption by radioisotope techniques and the measurement of gastrointestinal protein loss by means of (1) ^{131}I-labeled protein and polyvinylpyrrolidone, (2) ^{51}Cr-labeled albumin, and (3) ^{67}Cu-labeled ceruloplasmin. The last session, Clinical Applications of Radioisotope Techniques for the Study of Protein Metabolism, included studies of protein metabolism with labeled proteins and protein-like substances in relation to nutrition and in digestive diseases; selective permeability of the human placenta to plasma proteins and protein hormones; the total clearance of different preparations of gamma globulin; and the metabolism of gamma globulin in agammaglobulinemia and hypogammaglobulinemia.

(Joan E. Carden)

**Radioisotope Techniques in the Study of Protein Metabolism*, Technical Reports Series, No. 45, International Atomic Energy Agency, Vienna, 1965 (STI/DOC/10/45).

Section V

Isotopes and Radiation Technology

Safety

For some time Oak Ridge National Laboratory (ORNL) and others have been engaged in the testing of shipping containers and sealed sources.[1] More recently[2] the Isotopes Development Center program at ORNL has emphasized the testing of numerous sources, commercially available and otherwise, in order to set up a schedule of source-testing criterions to be used as guidelines for preparing, testing, and evaluating radioisotope sources, with particular emphasis on sealed sources. Working closely with ORNL is Sectional Committee N-5.4 (Use & Handling of Radioisotopes) of the United States of America Standards Institute and Laboratoire National d'Essais, Conservatoire National des Arts et Metiers, under contract to Centre d'Études Nucléaires de Saclay (France).

The early ORNL work involved the inspection and testing of many old sources, and, on the basis of the results, the criterions were established. Later work included further testing of these same sources and of additional prototype sources in order to evaluate the sources *and* the guidelines in terms of each other.

The purpose of this review is to indicate the status of the program and to delineate not only the various parameters involved but also some of the problems associated with establishing source criterions. Some testing information that resulted from specific requests by the AEC to evaluate particular sources is also included as matter of interest.

AEC—ORNL Source Safety Testing Program

By R. A. Robinson*

Abstract: The AEC—ORNL Source Safety Testing Program is designed to test a wide variety of sources and to set up a schedule of source-testing criterions to be used as guidelines for preparing, testing, and evaluating radioisotope sources.

Test-Procedure Development

There are many procedures for testing the containment integrity of radioactive sources, particularly in the areas of quality control, and for periodic testing of sources in use. The investigations at ORNL were designed to select those tests which would be useful in a source-classification system and which would provide more knowledge on the reliability and applicability of the various tests in the broader areas of source testing. Where existing testing techniques were considered unsuitable for source classification, modifications were made or new test procedures were developed.

The tests selected for use in the ORNL Source-Capsule Classification System were chosen because they are simple to perform, provide reliable, reproducible results, and, for the most part, require inexpensive test equipment. In each case, any failure resulting from the treatment is discerned by subjecting the source to the vacuum leak test. The general considerations for source evaluation are described below, and the rationale for selecting individual values within the evaluation categories is included. Tables 1 and 2 give the characteristics of the system.

LEAK TESTS

Since the ultimate basis for evaluating response to either temperature or structural tests is whether or not the source leaks, considerable time was spent in considering various leak tests that might be used. These are described for purposes of comparison. However, within its range of sensitivity, the vacuum leak test is the most reliable of the tests that were compared.

Vacuum Leak Test.[3] The source to be tested is immersed in ethylene glycol or isopropyl alcohol (chosen because of their low vapor pressures and surface tensions), and the pressure above the liquid is reduced to ~125 mm Hg absolute. A leak is indicated by a stream of bubbles rising through the

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liquid. The sensitivity of the test was determined by preparing a number of calibrated leaks, measuring the hole sizes microscopically, and measuring the leak rates. Under the test conditions described, leaks as small as 3×10^{-6} standard cm^3/sec can be readily detected, and leaks of 1×10^{-7} standard cm^3/sec have been detected by subjecting the capsule to a higher gas pressure immediately before the test.

The vacuum leak test can be used only if there is sufficient free volume in the capsule to support a stream of air bubbles in the event of a leak; 0.1 cm^3 is considered the minimum volume that can be used.

In cases where the free space is insufficient, a small amount of an easily soluble cesium or lithium salt (e.g., the chloride) is added to the nonradioactive source compound in the prototype capsule being tested. The source is alternately evacuated and vented underwater for a period of 20 min, and the water is analyzed flame photometrically.[4] If a soluble radioactive tracer ($^{137}CsCl$, $^{24}NaCl$) is used, standard counting methods can be employed for analysis.

Smear Test. The source is thoroughly wiped on all accessible surfaces, using moderate pressure, with a 25-mm-diameter absorbent paper disk, and the disk is then counted with an ordinary beta–gamma counter.

Hot-Water Bubble Test. The source at room temperature is quickly immersed in water at ~200°F. A stream of bubbles emanating from the source indicates a leak.

Air-Pressure Bubble Test. In a variation of the previous test, the source is placed in a pressure vessel at 540 cm Hg for 15 min, then quickly transferred to the hot water. A stream of bubbles indicates a leak.

Weight-Gain Test. The source, weighed to an accuracy of at least 0.01 g, is placed in a water-filled pressure vessel at 1800 cm Hg for 1 hr. A gain in weight indicates a leak.

Test with Helium or ^{85}Kr. Welded or brazed capsules may be tested for leaks by incorporating helium in the capsule during sealing. Any helium that leaks out can be detected with a helium mass-spectrometer leak detector.[5] If ^{85}Kr is used,[6] it is detected by counting.

Although the equipment required for helium or ^{85}Kr tests is quite expensive, the procedures are simple, readily adaptable to remote operations, and very sensitive.

Conclusions. The value of a thorough visual examination should not be discounted. In many instances examination of seal areas under moderate magnification (5× to 20×) clearly reveals porosity.

The vacuum leak test was the most sensitive and most reliable of the first five tests and detected leaks when all other methods failed. The test is not effective, however, when the free volume inside the source is too small to support a stream of bubbles (e.g., in interstitial therapy needles). This test also may not be satisfactory for a sizable leak from which the air may escape unnoticed in one or two large bubbles.

An evaluation is presently being carried out on the effect different sizes of leaks have on the amount of radioactivity that escapes. The high sensitivity of the helium and ^{85}Kr leak tests apparently is not sufficient to justify the high cost of equipment and the additional time for analysis.

MAXIMUM-TEMPERATURE TEST

The maximum-temperature test of the classification system is designed to determine the maximum temperature to which a source could be subjected for a short period of time. Fire hazard was of particular concern in this test.

The test is conducted in a commercially available laboratory-type furnace, which must be large enough to allow air circulation through the chamber and around the source in order to maximize oxidation of the source capsule during the test.

The maximum-temperature resistance category is intended to assure containment at high temperatures for short periods of time under accident conditions. The lowest limit, 400°F, is slightly above the temperature associated with photoflood lamps, steam sterilizers, and protective grills and cases around room heaters. It is felt that sources which cannot maintain containment for a short period of time at 400°F would be subject to failure from so many extraneous conditions at the point of use that they should be evaluated individually according to the type and quantity of activity. The cutoff temperatures for each class are set as follows:

Class A	400°F	—slightly below melting point of most soft solders; above the softening point of most present-day plastics
Class B	750°F	—below melting point of brazing and silver solders; above melting point of soft solders
Class C	1700°F	—equivalent to a normal 1-hr fire
Class D	2400°F	—short-term temperature limit for materials other than some superalloys and coated refractories
Class E	4500°F	—superalloy or coated-refractory-metal construction required

OPERATING-TEMPERATURE TEST

The operating-temperature test subjects a source to a temperature range that might reasonably be expected during usage. The test is conducted in the same type of furnace used for the maximum-temperature test.

Although the operating-temperature classification is not based on any specific applications, the various ranges involve reasonable situations. The lowest (Class A) range was chosen because it represents

a temperature span which is readily obtainable and over which any source might reasonably be expected to be used. The succeeding classes represent sources which are progressively more reliable from the standpoint of safety—up to those which can withstand the severest sustained conditions presently envisioned. One type of limiting application or environment is given with the temperature span of each class:

Class A	32 to 212°F	ice–water mixture to boiling water
Class B	−40 to 212°F	lowest long-term natural-environment temperature
Class C	−320 to 500°F	cryogenic to vacuum-system-"bakeout" temperature
Class D	−70 to 1700°F	conditions outside high-performance aircraft; molten-metal gaging
Class E	32 to 2400°F	suggested operating temperature of some power-conversion sources

THERMAL-SHOCK TEST

The thermal-shock test requirements also anticipate conditions to which a source might be subjected during use. The test development involved heating a source and then quenching it in any one of a variety of "fluids" at low temperature (the particular fluid or method used is optional as long as the required temperatures are obtained): air (room temperature), ice–water (32°F), ice–saltwater mixtures (0 to 32°F), liquid nitrogen (−320°F), trichloroethylene–Dry Ice mixtures (−70°F), or air–Dry Ice (−40°F). After being quenched, the source was transferred to an air atmosphere at −70°F, obtained by packing a container in a Dry Ice–trichloroethylene mixture.

The −70°F temperature is included to cover air transport and is that temperature normally experienced at an altitude of 40 thousand ft. The other values are intended to include the operating-temperature ranges for the individual classes. In Class E the thermal shock is extended beyond the operating range to include a source falling into a cryogenic storage vessel. The ranges for the classes are shown in Table 1.

TEMPERATURE–HUMIDITY TEST

The temperature–humidity test is conducted in any chamber capable of being regulated to reproduce and maintain the cycling requirements of temperature and relative humidity (RH) during the test: from 200°F at 95% RH to 35°F at 5% RH, with an 8-hr cycle time for a 48-hr period.

The temperatures and humidities chosen for this requirement represent the maximum and minimum temperature–humidity values likely to be experienced in actual source usage. In the ORNL test program, the only sources affected by this test failed within 48 hr; so this time limit was arbitrarily chosen. The temperature–humidity test requirement was considered unnecessary and was dropped from Classes C, D, and E because the other test requirements of these classes require a capsule construction that will not be affected by humidity conditions. Only one requirement is used for both Classes A and B.

PRESSURE TEST

Early work on pressure tests was performed using metallic sources that had a small tube welded into the capsule so that internal hydraulic pressure tests could be performed. This method was time-consuming, often weakened the capsules, and could not be readily used except on metallic capsules. For these reasons an external pressure test (Fig. 1) was selected for use in the classification system.

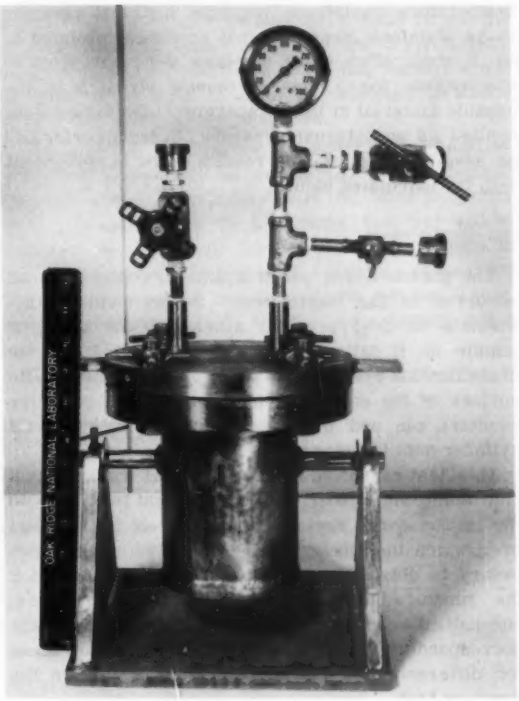

Fig. 1 External-pressure-resistance test.

This test is designed for those source capsules which fail the maximum-temperature test for some reason other than the internal pressure generated during the test—e.g., melting of a solder joint. This requirement also provides a means for further evaluation of a source which might be in a low-temperature class because of some test other than the maximum-temperature test but which is actually capable of withstanding a large internal pressure.

In addition to evaluating the external-pressure resistance of a source capsule, the test results can also be interpreted to give an indication of the internal pressure that the capsule could withstand. Normally the internal-pressure resistance is indicated by the maximum-temperature test results since the pressure in the capsule increases during this test owing to expansion of the gas in the void volume and volatilization of the source compound. However, in some instances a capsule fails to get into a particular temperature class because it fails some test other than the maximum-temperature test or it fails the maximum-temperature test for some reason other than internal pressure. To provide an indication of what the internal-pressure resistance would have been, the external-pressure requirements of the various classes were based on a calculated internal pressure for a capsule at the maximum-temperature resistance (capsule material assumed to be stainless steel, original pressure assumed to be 1 atm). After these stresses were corrected to compensate for a loss of tensile strength in the capsule material at this temperature, the values were applied as an external pressure. In tests performed on several capsules, the results were in agreement with the calculated values.

PUNCTURE TEST

The puncture test posed a difficulty that also had occurred in the impact test—it was virtually impossible to drop a small source without having it tumble as it fell. The procedure adopted for the classification system is to place the most susceptible surface of the source on top of the $\frac{1}{8}$-in.-diameter puncture pin and then to drop a free-falling metal cylinder onto the source.

This test represents a source falling from a height onto some small tool or stud. The test is similar to the impact-force resistance test except for the area over which the energy is absorbed. The heights necessary to obtain the requisite free-fall velocity for the puncture test are, with only two exceptions, one-half the heights required for free fall in the corresponding impact test. These lower values allow for differences in the mechanism of failure in the puncture test.

CRUSHING-FORCE TEST

The crushing test is performed by placing the source in a press having flat, parallel pressing surfaces. The source is oriented in the position most susceptible to failure (Fig. 2).

This test simulates the crushing force that would be applied if a weight were accidentally applied to the capsule. In the lower class this is equivalent to a 200-lb man stepping on the capsule; at higher classes it represents the crushing force of industrial vehicles or heavy objects on the capsule.

SHEAR TEST

The shear test uses a set of shear blocks with 90° shear edges, hardened to Rockwell C-65. One block is mounted solidly and holds the source in a fixed position. The shear force is applied to the other block, which is free to move downward in the direction of shear. The shearing edges are made so that they conform to capsule geometry.

In order to be consistent with the relationship of shear strength to tensile and compressive strength, the values for resistance to shear were taken as 50% of the requirements for resistance to crushing forces. A typical shear failure could occur in a teletherapy unit if the capsule became dislodged and the shutter were closed on it.

IMPACT TEST

In the impact test (Fig. 3), the source is placed on an essentially unyielding test pad with the most sus-

Figure 2

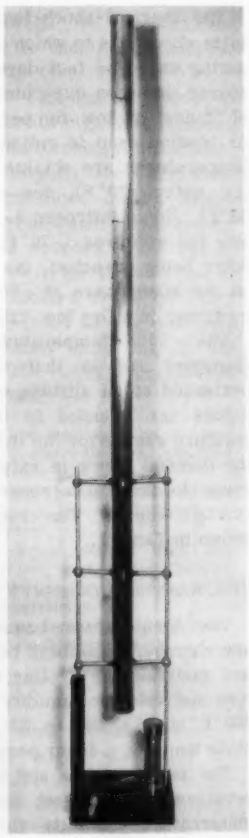

Figure 3

Fig. 2 Crushing-force test (same machine also used for shear testing).
Fig. 3 Device for impact test.

ceptible surface up. A 2-in.-diameter cylindrical weight is dropped vertically in free fall through a loosely fitting tube, which guides it and prevents it from tumbling as it falls; thus the flat end strikes the most susceptible surface of the capsule. A calibrated air-driven ram has also been used to supply the impact force.

The impact-force resistance test is intended to ensure that source capsules can withstand impacts resulting from accidents in which the capsule falls from a height to an essentially unyielding object. The lowest value simulates a fall from a storage shelf. Intermediate values refer to falls from various heights in buildings that might be gutted by fire or to falls from open industrial-type construction. The highest value represents a fall from a gantry.

VIBRATION TEST

The requirements for vibration resistance are taken from an Air Force test for aircraft and missile parts (Air Force Specification Mil-E-5272C). No tests are required for Classes III and IV because source capsules that normally fall into these classes are of rugged construction and are not normally subject to damage from vibration.

ABRASION TEST

Abrasion tests were performed on aluminum, brass, and stainless-steel rods ($\frac{3}{8}$ in. in diameter by 1 in. long, to simulate a typical source-capsule shape). The rods were held at a 45° angle against a rotating disk covered with 180-grit abrasive paper. The pressure of the metal against the abrasive was varied by attaching weights to the rod. The amounts of material removed by abrasion were so small that source-capsule failure from this cause was not considered likely, and the abrasion requirements were eliminated from the classification system.

CHARACTERISTICS OF ORNL SOURCE-CAPSULE CLASSIFICATION SYSTEM

Tables 1 and 2 summarize the source-classification information. The classification system comprises two sections: temperature classes A through E and structural classes I through V. Each capsule design under consideration will be classified in both these categories to give a composite classification (e.g., I-C or III-A), which describes both the temperature and the structural integrity of the capsule.

For the initial classification of a source capsule, a study of the capsule design will usually reveal the most susceptible areas of failure due to mechanical and temperature limitations. Tests may be performed in these areas first in order to establish the limits in the most likely areas of failure.

For reasons of economy the source-capsule classification tests may be performed in any desired sequence. Source capsules may be used for more than one test if it is not likely that they will be damaged to an extent that would cause failure in additional testing; however, if a capsule is used for more than one test and it fails the additional test, the test failure shall be verified by retesting with a new capsule.

Table 1 ORNL SOURCE-CAPSULE CLASSIFICATION SYSTEM: TEMPERATURE CHARACTERISTICS

Class	Maximum-temperature resistance, °F	Operating temperature, °F	Thermal shock, °F	Temperature–humidity cycling
A	400 for 1 hr	32 to 212	212 to 32, then to −70	200°F–95% RH to 35°F–5% RH; 8-hr cycle time for 48-hr period
B	750 for 1 hr	−40 to 212	212 to 0, then to −70	Same as A
C	1700 for 1 hr	−320 to 500	500 to 32, then to −70	No requirement
D	2400 for 1 hr	−70 to 1700	1700 to −70	No requirement
E	4500 for 30 min, 2400 for 1 hr	32 to 2400	2400 to −320	No requirement

Table 2 ORNL SOURCE-CAPSULE CLASSIFICATION SYSTEM: STRUCTURAL CHARACTERISTICS

Class	External pressure, psig	Puncture resistance, ft/sec onto $\frac{1}{8}$-in.-diameter pin	Crushing force, lb for 1 hr	Shear strength, lb for 1 hr	Impact force (< 2 lb source wt.) force, ft-lb	Impact force (> 2 lb source wt.) free fall, ft	Vibration
I	30	14	200 for 15 min	40 for 30 min	20	10	10 cps at 10 *g*'s (longest axis) for 12 hr
II	100	23	200	100	30	15	Same as I
III	1,000	31	2,000	1,000	60	30	No requirement
IV	3,000	44	20,000	10,000	120	60	No requirement
V	10,000	62	50,000	25,000	240	120	4 hr at each resonance*

*5 to 500 cps and 0.001- to 0.1-in. double amplitude applied along each of three perpendicular axes; when no resonant frequencies are found, vibrate for 12 hr at 0.018-in. double amplitude and 150 cps on each axis.

The selection of individual values in the ORNL Source-Capsule Classification System is directly related to the excellent safety record associated with domestically manufactured sealed sources. The early work in the ORNL source-testing program was designed to define those characteristics of commercial source capsules which were responsible for the established safety record. The results of these investigations and other tests strongly influenced the selection of most of the values in the source-capsule classification tables. The classification tests do, therefore, reflect current safe practice in sealed-source design.

Specific reasons in terms of particular applications or accident possibilities could not be given for all values selected for the tables. In some instances the values were established as logical extrapolations of conditions for which specific reasons were given. The intention was to set parameters for each successive class that would represent a consistent progression of containment-related characteristics toward greater reliability under progressively more adverse conditions. The values are arranged so that essentially all domestically manufactured sealed sources will fit somewhere in the classification system.

Examination of Old Sources

Of considerable interest to the source-testing program was the examination of old sources, some of which were still integral and others of which had been withdrawn from service because of leaks. Several of these are described briefly.

CESIUM TELETHERAPY SOURCE

A 9-year-old ^{137}CsCl source (1540 curies) was examined[7] to determine the effects of aging on stainless-steel capsules for radioisotope sources. It contained two ^{137}CsCl pellets encapsulated in two concentric type-316 stainless-steel containers (Fig. 4). The inner capsule was plugged with a 316 stainless-steel plug ~1.5 in. long, and the outer capsule was plugged with a threaded plug of 410 stainless steel. Both plugs were sealed with silver solder.

The source was visually inspected upon receipt by ORNL. The outer capsule appeared to be in excellent condition; no corrosion or discoloration could be detected.

The source was tested for leaks by immersion in ethylene glycol in a glass vacuum vessel at a pressure of ~5 in. Hg absolute; no leaks were detected.

Visual examination of the exterior of the inner capsule revealed no evidence of corrosion or deterioration. After the inner capsule of the source had been opened and the pellets had been removed, it was observed that the upper edge of one pellet and the adjacent area on the capsule were the only affected (discolored) areas. The window and the inside of the capsule were bright and shiny. Inspection under 80× magnification did not reveal any pitting or cracking of the inside surfaces of the capsule or the window. The capsule was then decontaminated by ultrasonic cleaning; metallurgical examination indicated no change in the crystalline structure and no deterioration of the capsule wall or window.

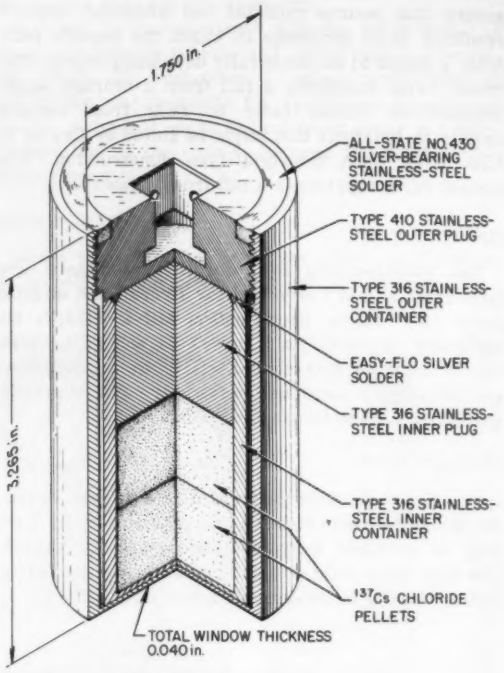
1.750 in.
ALL-STATE NO. 430 SILVER-BEARING STAINLESS-STEEL SOLDER
TYPE 410 STAINLESS-STEEL OUTER PLUG
TYPE 316 STAINLESS-STEEL OUTER CONTAINER
EASY-FLO SILVER SOLDER
TYPE 316 STAINLESS-STEEL INNER PLUG
TYPE 316 STAINLESS-STEEL INNER CONTAINER
^{137}Cs CHLORIDE PELLETS
3.255 in.
TOTAL WINDOW THICKNESS 0.040 in.

Fig. 4 Nine-year-old ^{137}Cs source.

U. S. NAVY CALIBRATION SOURCES

A number of small U. S. Navy calibration sources that had been removed from service because of excessive transferable activity found during periodic smear tests were examined and found to leak. In most cases these leaks appeared to have resulted from incomplete soldering during manufacture.

BETA-GAGE SOURCES

Five beta-gage sources that had withstood a factory fire were examined.[8] Each capsule was a ½-in.-diameter by ¼-in.-high right-circular cylinder with a 1.5-mil-thick window brazed to the body of the source. Each source had been prepared by depositing 10 to 12 mc of ^{90}Sr on a ceramic disk, covering the radioactivity with a silicate, and baking the unit. The ceramic disk was then placed in the capsule with the radioactive end adjacent to the thin window, and the capsule was closed with a threaded plug sealed into place with silver solder.

One of the sources, when cleaned with weak nitric acid and a soft wire brush, regained most of its original luster. The solder on the threaded plug closure had melted off in the fire, and a small leak was detected through the threads. The window was slightly corroded but did not leak. The second source also regained its luster, and very little of the solder had melted, although the window, which had buckled and cracked, did leak. The window of the third source was also buckled and appeared cracked, but it did not leak in the vacuum leak test. About half of the solder had melted off the threaded plug, and the source appeared to leak momentarily when subjected to the leak test. It was concluded, however, that the leak resulted from air trapped in the threads. The fourth source was embedded in a large piece of aluminum that had resolidified. The retainer plug from the device was fused to the threaded plug of the source. When cleaned, the source had a fine crack that extended around its circumference. The window was badly oxidized, and parts of it were missing. The fifth source, still in the shutter mechanism of the gaging device, was somewhat distorted from the fire. There was no evidence of leakage, however, and close visual examination did not reveal any damage.

This particular study indicated that, even where leakage had occurred, most of the activity remained within the source—a fact which emphasizes the importance of the form of the source material and points up the importance of source-form consideration in the application-rating guide.

^{60}Co TELETHERAPY SOURCES

Eighteen lead O-ring ^{60}Co teletherapy sources from 5 to 8 years old were tested to determine the effects of age. The source capsules (Fig. 5) had five parts: body, plug, cup, and two lead O-rings; the body and plug were constructed of an alloy containing 90% tungsten, 6% nickel, and 4% copper.

Three types of sources were involved, differing principally in the type of plug used. One plug was designed to hold 2-cm-diameter cobalt wafers; a second could hold six 1- by 1-cm pellets in a "cloverleaf" array;[9] the depths of both of these plugs could be varied. The third, 0.542 in. deep, was used for 1- by 1-mm cobalt pellets. The outer seal ring was a $\frac{1}{8}$-in.-diameter antimony-free lead wire formed into an O-ring of $1\frac{3}{8}$ in. inside diameter.

The cup was made of 0.020-in.-thick type-305 stainless steel with an inside diameter of 1.187 in. and an inner height of 1.306 in. The cup had an open top with a 1.5-in.-diameter lip. The cup was seamless, annealed at 2000°F, and air cooled before and after being formed. The lip on the open top of the cup was the second, or inner, sealing surface. The seal ring was a $\frac{1}{16}$-in.-diameter antimony-free lead O-ring of $1\frac{3}{16}$ in. inside diameter.

Visual inspection indicated that the capsules were not damaged. They were then subjected to several of the leak tests already outlined:

a. Smearing with absorbent paper to determine the presence of transferable activity
b. Weighing, immersing in water at 300 psi for 1 hr, and reweighing
c. Resmearing with absorbent paper to determine the presence of transferable activity
d. Placing in air at a pressure of 90 psig for 15 min and then in hot water and observing for bubble formation
e. Smearing for a third time with absorbent paper for transferable activity
f. Immersing in ethylene glycol and decreasing the pressure to 5 in. Hg absolute

The results in Table 3 show that the vacuum leak test (*f*) gave definite indications of leaks in all cases.

When disassembled, each source showed breaks in both O-ring seals, with spaces of $\sim\frac{1}{16}$ in. between the ends. The seals appeared to have been pulled apart in the initial sealing process. No other damage was observed. Although all 18 sources were stored in the same container, there was no evidence of melted gaskets; and, when water was put into them, no steam arose although they dried rapidly.

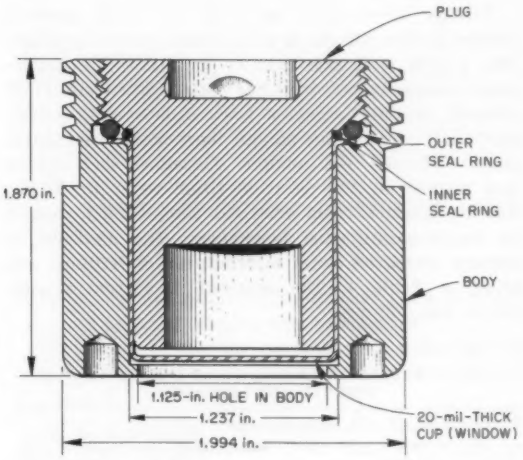

Fig. 5 Cobalt-60 teletherapy source.

Four of the sources were reassembled without the cobalt and subjected to structural tests to determine the maximum physical conditions a source of this type could withstand. The first unit was dropped onto a 0.25-in.-diameter rod with the 20-mil-thick window of the source striking the end of the rod. The first drop was from 6 in., and subsequent drops were made in 6-in. increments up to 2 ft, at which height the window ruptured. The second and third sources

Table 3 COBALT-60 TELETHERAPY-SOURCE TESTS

Source No.	(a) Smear,* dis/min	(b) Wt. gain, g	(c) Smear,* dis/min	(d) Bubbles	(e) Smear,* dis/min	(f) Bubbles
1	60	0.75	10	None	13,570	
2	600	−0.90	2,300	Few	9,890	
3	60	0.85	10	None	42,070	
4	1,750	0.05	406,180	None	727,630	
5	6,000	1.56	12,700	None	109,340	
6	27,700	0.90	147,300	Many	35,550	
7	12,674	−0.55	155,180	Many	102,000	
8	250	0.19	Background	None	105,800	
9	2,000	0.95	19,800	None	18,430	
10	8,960	2.15	270,140	Many	552,140	
11	8,310	2.70	16,000	Few	377,820	
12	17,500	1.85	8,600	Few	142,780	
13	143,540	0.10	77,120	Many	23,810	
14	203	2.75	155,780	Small for 10 sec	124,850	
15	5,470	2.82	7,140	None	141,110	
16	39,420	−0.90	51,090	Many	22,760	
17	6,330	0.75	310,300	None	298,420	
18	970	2.00	99,600	Many	178,460	

Bubbles in *all* cases

*Smears taken of total source area.

were dropped initially from a height of 2 ft, and in both cases the window ruptured. The fourth capsule was placed on its side in a 50-ton press, and force was applied until the body cracked at a total applied force of 84,000 lb.

U. S. ARMY TRITIUM LEVEL VIALS

Tritium level vials, used by the U. S. Army to determine the attitude of army tanks, were examined. The smear technique used by the Army was duplicated (smear taken with filter paper wetted with ethyl alcohol), and the samples were counted with a proportional counter. In all but four cases, the initial smear "blocked" the counter. Therefore each source was soaked in 100 ml of distilled water for 24 hr, and the solution from each was sampled and counted by liquid-scintillation techniques. The amounts of activity removed (total microcuries of tritium) are given in Table 4. The nominal ^3H content of each device was 100 mc.

Table 4 RESULTS OF INITIAL SMEAR AND WATER-SOAK TESTS ON ARMY SOURCES

Source	Initial wipe, dis/min	Activity removed by water soak, μc	Second wipe, dis/min	
			G–M tube	Proportional
S-H-99A	*	148	Background	17,024
S-H-99B	34,241	865	Background	18,752
S-H-99C	*	1,257	Background	11,598
S-H-99D	*	1,131	Background	23,040
S-H-99E	*	878	Background	15,784
S-H-99F	78,120	995	Background	37,376
S-H-99G	36,032	608	Background	10,750
S-H-99H	*	851	Background	31,936
S-H-99I	4,400	622	Background	14,376
S-H-99J	*	1,077	Background	19,648
S-H-99K	*	1,099	Background	12,944
S-H-99L	*	This source was disassembled; therefore only the initial wipe test was performed.		

*Blocked counter—probably >100,000 dis/min.

The device was quite similar to an ordinary carpenter's level. The source consisted of a glass vial containing a liquid, resting in an aluminum trough containing a tritiated phosphor. The trough and vial were held together by a brass outer casing and plaster of paris. The phosphor appeared to be adhering tightly to the aluminum, and there was no loose surface material. The origin of the activity is not entirely certain unless it was from radiolysis of the tritiated polymer, although this has not been a serious problem with most watch-dial paints; however, approximately thirty times as much active material is in these sources as in a comparable volume of watch-dial paint.

Air at room temperature was passed across several sources for 1 hr. The air was scrubbed with toluene and water and sampled to determine the amount of tritium removed in the 1-hr period. The activity removed varied from 2.5 to 82 μc. An opened source was exposed to a vacuum of 40 μ Hg; the total amount of activity released during 1 month was ~12 μc.

U. S. ARMY LENSATIC COMPASSES

A set of eight U. S. Army lensatic compasses, each containing 75 mc of tritiated phosphor paint, was subjected to smear, water-leach, and temperature–humidity tests. The smear tests revealed moderate contamination (608 to 29,000 dis/min) of the compass surfaces and carrying cases. Smears of exposed paint areas showed these areas to be highly contaminated (up to 546,000 dis/min). The amount of transferable activity did not change appreciably as a result of exposure to a temperature–humidity test. Compasses exposed to the standard 24-hr water-leach test lost an average of 5.66×10^7 dis/min or 0.29% of the initial activity.

Testing of Commercial Sources

Also of interest to the program was the testing of off-the-shelf sources purchased from commercial vendors. The results of several of these tests follow.

MEDICAL SOURCES

Interstitial Therapy Needles. A special series of tests was used to determine the structural strength of interstitial therapy needles (Fig. 6). The impact

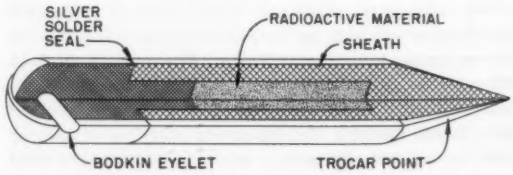

Fig. 6 Cobalt-60 needle.

test consisted in dropping weights onto the center of the needle which was supported on each end (Fig. 7*a*). Bend tests were carried out by suspending weights from the tip of the needle, which was clamped in a vise at a specific position from its point end (Fig. 7*b*). The needle points were also subjected to a break test (Fig. 7*c*) by holding the needle in a vise just behind the seal formed by the point and suspending weights from the opposite end.

In the tensile test (Fig. 8*a*), each end of the needle was secured in a clamping block, and opposing forces were applied to the blocks. The shear test (Fig. 8*b*) was similar to the resistance-to-shear test of the classification system—one end of the needle was securely held and a shearing block moved against the needle.

For corrosion tests one needle was implanted in a rabbit, and other needles were tested by immersion in blood plasma. Additional tests were made to determine the effect of sterilization procedures.

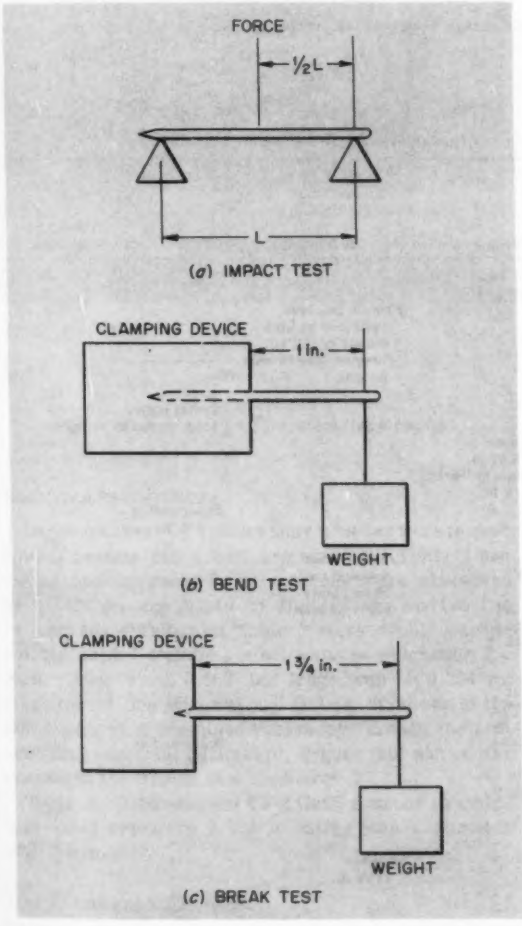

Fig. 7 Structural tests for interstitial therapy needles.

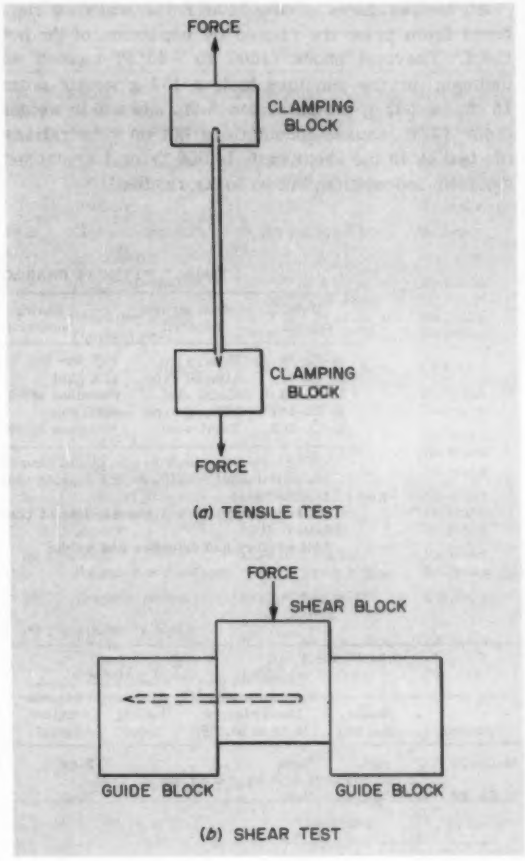

Fig. 8 Tensile and shear tests for interstitial therapy needles.

Five types of therapy needles were submitted for testing; their physical characteristics are detailed in Table 5.

Table 6 summarizes some of the tests made on these needles. Additional tests made on wires removed from the needles are covered in the original report[2] but are not tabulated here.

TELETHERAPY SOURCES

Cesium. This type of source contained CsCl doubly encapsulated in stainless-steel cylinders with welded closures. A 0.020-in.-thick window was machined in one end of each capsule, providing a total window thickness of 0.040 in. Each assembly was 1.5 in. in diameter by 1.5 in. high and weighed ~195 g. Thirteen samples of this type of source were tested (nine in the first series of tests, four in the second).

At temperatures above 1400°F the windows ruptured from pressure caused by expansion of the hot CsCl. Thermal shock (1000 to −40°F) caused no damage; in the puncture test, a 194-g weight from 15 ft, a 582-g weight from 5 ft, and a 6-lb weight from 12 ft caused indentations but no penetrations (no leaks). In the shear test, 10,000 lb for 1 hr caused a 0.1-in. indentation, but no leaks resulted.

The second set was tested to permit classification under the ORNL Source-Capsule Classification System and verification of the results of the tests on the first set. The source type was classified II-B on the basis of the tests.

Cobalt. Doubly encapsulated welded stainless-steel cylinders ($1^{7}/_{16}$ by $1^{3}/_{32}$ in.) containing cobalt pellets

Table 5 PHYSICAL CHARACTERISTICS OF THERAPY NEEDLES

Test sample	Cobalt source material	Casing material	Length, cm	Outside diameter, cm	Wall thickness, mm
M-Co-3*	Metal wire	80% Pt–20% Ir alloy	2.7	0.165	0.30
M-Co-4†	Alloyed wire	10K gold	4.0	0.17	0.25
M-Co-6‡	Metal wire	Stainless steel	4.0	1.06	0.19
M-Co-14§	Alloyed wire	9.5K gold	4.0	-	0.18
M-Co-20¶	Metal wire	Stainless steel	1.8	0.127	0.10

*Silver-soldered bodkin-type eyelet formed the capsule seal.
†Soldered point constituted the capsule seal.
‡Welded seal.
§Silver-soldered stainless-steel point constituted capsule seal; had removable stainless-steel eyelet.
¶All sources had defective end welds.

Table 6 SUMMARY OF TESTS ON THERAPY NEEDLES

Source	Smear, dis/min	Corrosion test (contamination of blood plasma, 16 hr at 98.6°F)	Leak test: 6 hr in boiling water	Leak test: Vacuum ethylene glycol	Bend test	Impact test (60-g ball dropped onto center of needle)	Tensile test	Shear test severed, lb
M-Co-3A	40	None	-	None	Intact when bent to 45°	-	-	-
M-Co-3B	30	None	-	None	-	From 6 in.; leak indicated by high smear count, but vacuum test showed no leak	-	-
M-Co-3-1A	-	-	-	-	-	-	-	155
M-Co-3-1C	-	-	-	-	-	-	Eyelet pulled out at 195 lb	-
M-Co-4	None	37 dis/(min)(ml)	None	-	Split open at 8.07 lb	-	-	-
M-Co-4-1A	-	-	-	Leaked badly	Permanent "set" at 18 lb	-	-	-
M-Co-4-1B	-	-	-	-	-	-	Separated at 210 lb	-
M-Co-4-1C	-	-	-	-	-	-	-	157
M-Co-6	None	46 dis/(min)(ml)	-	-	-	Cracked when dropped from 36 in.; however, no leak	-	-
M-Co-6-1A	-	-	-	None	Bent to 20° at 12 lb	-	-	-
M-Co-6-1B	-	-	-	-	-	-	-	205
M-Co-6-1C	-	-	-	None	-	-	-	-
M-Co-14*	None	-	None	None	Split at 4.77 lb	-	-	-
M-Co-14-1A	-	-	-	None	-	-	-	175
Mo-Co-14-1B	-	-	-	None	-	-	Separated at 238 lb	180
M-Co-14-1C	-	-	-	None	Bent to 20° at 22 lb	-	-	207
M-Co-14-1D	-	-	-	Leaked at eyelet	-	-	-	356
M-Co-14-1E†	-	-	-	None	-	-	-	-
M-Co-20A	-	-	-	-	-	No leaks at 24 in.	-	-
M-Co-20B	-	-	-	-	-	-	-	190
M-Co-20C	-	0.13 ppm Co	-	-	-	-	-	-

*Insertion in lower intestinal region of rabbit for 1 week resulted in no measurable contamination.
†Subjected to temperature–humidity test. No damage noted.

and weighing 198 g were classified III-C after testing. Figure 9 shows the results of the impact test on one of the sources.

Fig. 9 Impact test on ^{60}Co teletherapy source.

RADIOGRAPHY SOURCES

Large numbers of radiography sources were tested; typical results for cobalt are shown in Table 7 and for cesium in Table 8. All sources were classified as III-C on the basis of the test results. The 11 sources detailed in Table 7 were doubly encapsulated welded stainless-steel sources weighing ~2 g each. They were 0.680 in. long, with a 0.264-in. diameter at one end and a 0.203-in. diameter at the other. Source A contained radioactive cobalt; the rest contained inactive material. Figure 10 shows the results of the impact test on source 2C.

Table 8 deals with ten 42-g CsCl sources in stainless-steel cylinders 0.880 in. high, with a diameter of 0.738 in.

MISCELLANEOUS SOURCES

A number of additional sources of various kinds were investigated, including Po–Be neutron sources

Table 7 TEST RESULTS ON COBALT RADIOGRAPHY SOURCES

Source	Type of test	Conditions	Results
A	Smear	Filter paper	110 dis/min
	Vacuum leak test	---	Leaked
	External-pressure leak test	300 psi for 1 hr	No weight gain
	Hot-water bubble leak test	90 psi for 1 hr, then to 200°F	No leak indication
1A	Crushing	13,400 lb	No leaks
1B	External pressure	22,000 psi for 1 hr	No leaks
1C	Not tested		
2A	Temp. resistance	1700°F for 1 hr in air; −320°F for 24 hr	No leaks
	Operating temp.	500°F for 24 hr in air	No leaks
	Thermal shock	500 to 32 to −76°F for 15 min	No leaks
	Temp. resistance	2550°F for 1 hr in air	Cracks
2B	Impact	12 lb for 5 ft	No leaks
	Thermal shock	1700 to −76°F	No leaks
2C	Crushing	2000 lb for 1 hr	No leaks
	Shear	1000 lb for 1 hr	No leaks
	Puncture resistance	0.077 lb from 5 ft	No leaks
3A	External pressure	1000 psi for 15 min	No leaks
	Crushing	2000 lb for 1 hr	No leaks
	Impact	12 lb from 5 ft	No leaks
3B	Shear	1000 lb for 1 hr	No leaks
	Puncture resistance	0.022 lb from 5 ft	No leaks
3C	Thermal shock	500 to 32 to −76°F for 15 min	No leaks
3D	Temp. resistance	1700°F for 1 hr in air; −320°F for 24 hr	No leaks
	Operating temp.	500°F for 24 hr	No leaks

Table 8 TEST RESULTS FOR CESIUM SOURCES

Source	Type of test	Conditions	Results
1A	Temp. resistance	1700°F for 1 hr in air	No leaks
	Temp. resistance	2550°F for 10 min in air	No leaks
1A	Temp. resistance	1700°F for 1 hr in air	No leaks
	Operating temp.	750°F for 24 hr, then to −320°F for 24 hr	No leaks
	Thermal shock	500°F for 1 hr, to 32°F, then to −76°F for 15 min	No leaks
	Temp. resistance	2550°F for 1 hr	Leaked
1B	Impact	12 lb from 5 ft	No leaks
	Crushing	20,000 lb for 1 hr	Ruptured
1C	Crushing	2000 lb for 1 hr	No leaks
	Puncture	0.275 lb from 5 ft	No leaks
1D	Shear	1000 lb for 1 hr	No leaks
	Puncture resistance	0.556 lb from 5 ft	No leaks
	Puncture resistance	1.11 lb from 5 ft	Small leak
	Shear	24,000 lb	Failed
1E	Not tested		
1F	Not tested		
2A	External pressure	1000 psi for 1 hr	No leaks
	Puncture resistance	0.220 lb from 5 ft	No leaks
	Crushing	2000 lb for 1 hr	No leaks
	Shear	1000 lb for 1 hr	No leaks
2B	Operating temp.	−320°F for 24 hr	No leaks
	Operating temp.	500°F for 24 hr in air	No leaks
	Impact	12 lb from 5 ft	No leaks
2C	Thermal shock	500 to 32 to −76°F for 15 min	No leaks
	Temp. resistance	1700°F for 1 hr in air	No leaks

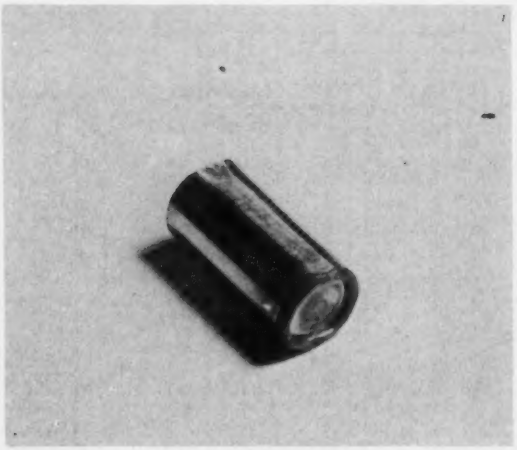

Fig. 10 Impact test on ^{60}Co radiography source.

Table 9 TEST RESULTS ON WELD-SEAL Po–Be NEUTRON SOURCE*

Test	Classification	Results
Maximum temp.	C	No damage, no leaks
Operating temp.	C	No damage, no leaks
Thermal shock	C	No damage, no leaks
Thermal shock	D	Microscopic examination showed no cracks in capsule or weld; no leaks
External pressure	III	No damage, no leaks
Puncture resistance	V	Scratched but no indentation; no leaks
Shear	III	Small indentation, no cracks; no leaks
Impact	III	Flattened from 0.625 to 0.611, no breaks in weld; no leaks

*1- by 0.625-in. stainless steel; 33 g. Given III-C classification.

Table 10 TEST RESULTS OF KRYPTON GAGE SOURCES*

Source	Type of test	Conditions	Results
3A	Puncture	0.051 lb from 4 ft	Punctured
3B	External pressure	100 psig for 1 hr	No leaks, but collapse at 85 psi
3C	Crushing	200 lb for 15 min	No leaks
	External pressure	30 psig for 1 hr	No leaks
	Puncture	0.027 lb from 3 ft	No leaks
	Shear	40 lb for 1 hr	No leaks
3D	Operating temp.	−76°F for 24 hr in air	No leaks
	Operating temp.	212°F for 24 hr in air	No leaks
	Temp. resistance	750°F for 1 hr in air	No leaks
	Thermal shock	212 to 0 to −76°F for 15 min	No leaks
	Impact	4 lb from 5 ft	Crushed

*1½- by ¾-in. stainless steel; 12 g. Classified I-B.

(Table 9); ^{85}Kr, ^{60}Co, and ^{90}Sr (beta and gamma) gages (Tables 10 and 11); several old ^{137}Cs calibration sources (all of which leaked); static-charge eliminators (Table 12); annular cesium sources containing CsCl powder doubly encapsulated in welded stainless steel; cylindrical rod (½ by 12 in.) sources containing cobalt pellets doubly encapsulated in welded stainless steel; CsCl sources doubly encapsulated in heliarc-welded aluminum; iridium in aluminum; thulium in stainless steel; cesium, singly encapsulated

Table 11 TEST RESULTS OF ^{60}Co GAGE SOURCES*

Source	Type of test	Conditions	Results
1A	Temp. resistance	1700°F for 1 hr in air	No damage
	Operating temp.	750°F for 24 hr	No damage
	Operating temp.	−320°F for 24 hr	No damage
	Thermal shock	500 to 32 to −76°F for 15 min	No leaks
	Temp. resistance	2550°F for 1 hr in air	Source melted
1B	Impact	12 lb from 5 ft	Cracked
1C	Crushing	2000 lb for 1 hr	Cracked
1D	Impact	6 lb from 5 ft	Cracked
1E	Crushing	200 lb for 1 hr	No leaks
	Impact	5 lb from 4 ft	Cracked
1F	Shear	100 lb for 1 hr	No leaks
	Puncture	0.044 lb from 30 in.	No leaks
	Puncture	0.088 lb from 5 ft	No leaks
	Thermal shock	1700 to −76°F	No leaks

*0.642- by 0.25-in. doubly encapsulated stainless steel; 3 g. On basis of these and other tests, classified II-C.

in stainless steel and sealed with silver solder; and $^{90}SrTiO_3$ sources. In addition, several types of prototype sources which are not in commercial production have been tested, of which the following are typical.

Sources for Portable X-Ray Unit. Twelve source models were tested, but all failed to meet the minimum performance (Class I-A) covered in the ORNL Source-Capsule Classification System. The sources were particularly susceptible to impact and thermal-shock failure. In addition, several samples gave substantial indication of leakage, both visually and analytically, during the initial leak test before any classification tests were performed. These prototype sources were sealed by means of a threaded plug which was not back-soldered or welded.

^{60}Co Irradiator Sources (BNL). These sources, ⅝- by 12- by 1/16-in. strips of cobalt, doubly encapsulated in welded stainless-steel containers (with overall dimensions of ¾ by 13 by 5/32 in.), are excellent in quality and workmanship; they met or exceeded all the test requirements for ORNL Class III-C.

^{90}Sr Beta-Source Coil. This welded stainless-steel coiled source (Fig. 11) was ⅛ in. in diameter and 6 ft long, with a 20-mil wall, and wound into a 2-in. coil. In general, it was about one class higher than the usual beta source having an equivalent window

Table 12 RESULTS OF TESTS ON ^{210}Po STATIC-ELIMINATION FOILS

Environmental test	Method of evaluation	Activity, dis/min			
		Sample S-Po-45A	Sample S-Po-45B	Sample S-Po-45C	Sample S-Po-45D
Control	Smear (moistened)	-	-	52	60
Control	Smear (wet)	130	0	26	216
Temperature–humidity (212°F, 100% RH; 32°F, 100% RH)	Smear	1484	-	-	-
Temperature–pressure (–76°F, 180 mm Hg; 140°F, 760 mm Hg)	Smear	-	42	-	-
Vibration (125 cps for 1 hr)	Smear	-	280	-	-
Vibration in water	Smear	-	2545	-	-
	Solution	-	20	-	-
Leach	Smear	-	0	-	-
(in water)	Solution	-	1479	-	-
(Kodak Acid Fixer)	Smear	-	0	-	-
	Solution	-	98	-	-
(Kodak Microdol X)	Smear	-	0	-	-
	Solution	-	2657	-	-
Elevated temperature	Smear	5647	-	-	21
	Filter	1544	-	-	226
	Cold trap	150	-	-	0
	Scrubber	250	-	-	0
Saliva	Smear	-	-	-	208
	Saliva	-	-	-	300
Bend	Bend 1 (smear)	-	-	-	2589
	Bend 2 (smear)	-	-	-	54
	Bend 3 (smear)	-	-	-	96
	Bend 4 (smear)	-	-	-	105
	Bend 5 (smear)	-	-	-	0
	Bend 6 (smear)	-	-	-	0
	Bend 7 (smear)	-	-	-	0
	Bend 8–16 (no smears taken)				
	Bend 17 (smear)	-	-	-	21
	Probe	-	-	-	7500
	Edge smear	-	-	-	0

Fig. 11 Coiled ^{90}Sr beta source.

thickness. It met all the Class C temperature tests and all the Class III structural tests except the shear test. (PSB)

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Section VI

Isotopes and Radiation Technology

Isotopes Around the World

Use of Large Radioisotope Sources in Canada, United Kingdom, and Japan*

By Martha Gerrard

Abstract: The ANS 1966 conference on large-scale production and applications of radioisotopes included a section on international programs in this field. Information from the conference on Belgian work on ^{227}Ac and French work on several nuclides was reviewed in Isotopes and Radiation Technology, *4(2), as were most of the other conference papers. In the current issue we present information from Canada, the United Kingdom, and Japan. Information on International Atomic Energy Agency work will be reviewed in a future issue of* Isotopes and Radiation Technology.

Canada

Cobalt-60, the most widely used nuclide for large-scale isotope irradiation applications, is produced in Canada in the NRX and NRU reactors at Chalk River. The NRX reactor began operation on July 22, 1947, and shipments of radioisotopes started 3 months later. In 1957 production facilities were increased with the startup of the NRU reactor. The target materials used for making ^{60}Co, selected after consideration of reactor economics, convenience in handling and encapsulation, and suitability for customer use, are cobalt metal slugs 1 in. long by 0.25 in. in diameter and cylindrical (1 mm in diameter by 1 mm high) nickel-plated pellets sandwiched between aluminum sheets.

The growth in use of radioisotopes is shown by the increase in number of licenses issued by the Atomic Energy Control Board of Canada, i.e., from 19 in 1949 to 1410 in 1964. With allowances for population differential, this growth is similar to that in the United States. The rate of growth was especially dramatic in the early years (Fig. 1).

Irradiation plants supplied by Atomic Energy of Canada Ltd. (AECL) have been designed using the concept of a machine housed completely in the irradiation cell. The radiation-resistant machine is built and pretested in the AECL plant. Experience indicates that this type of plant will be in operation about 95% of the time when operating on a 24-hr/day basis. Both wet and dry source storage have been used, but AECL now believes that wet storage is more flexible and safe, especially for large sources. The general design is an optimization of throughput, ef-

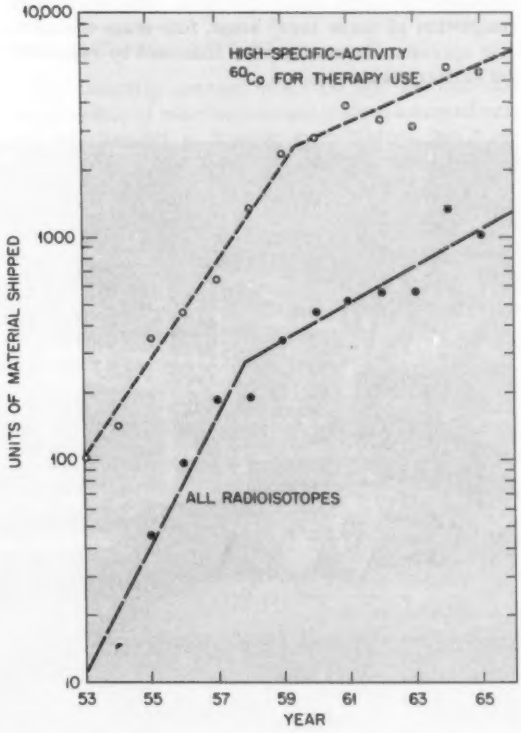

Fig. 1 Increase in AECL isotope shipments

*Based on papers by R. F. Errington, H. Sheard, and E. Munekata at the American Nuclear Society Topical Meeting in Augusta, Ga., 1966, printed in Large Scale Production and Applications of Radioisotopes, USAEC Report DP-1066, Vol. I.

ficiency, dose uniformity, size, and weight of the equipment. Some typical design parameters are:

	Early designs	Recent designs
Efficiency, %	10	45
Dose uniformity, %	±15	±15
Throughput, lb/hr	2,500	60,000
Plant capacity, curies	20,000	600,000

Large-scale ^{60}Co sources are used in Canada for food preservation, pharmaceutical supplies sterilization, and cancer teletherapy. AECL also designs and exports irradiation plants.

FOOD PRESERVATION

In food-preservation studies the same plan is followed for each food. First the minimum dose required to achieve a desired result and the effect of this dose on product quality and acceptability are determined. If this step appears promising, clearance is requested of the Food and Drug Directorate of the Department of National Health and Welfare, wholesomeness data from animal feeding trials being submitted. If clearance is obtained, pilot-plant studies are made to ensure that the experimental results are applicable to large-scale operation. After successful completion of these three steps, full-scale commercial operation is attempted if indicated by results of the earlier steps.

For the most economic utilization of funds and effort, Canadian work on radiation preservation was initially concentrated on one product—potatoes. This process has now been carried through to commercialization. The study of gamma irradiation of potatoes to inhibit sprouting was started in 1956. A cooperative program followed, in which seven federal and three provincial government departments, five universities, and a number of private industries participated. The results were Food and Drug Adminstration clearance for potatoes in 1960, followed by construction of a mobile irradiator in 1961 and of a commercial plant in 1965. The success of early experiments on potatoes was followed by experiments on onions, fruits, fish, eggs, and meat. It is thought that export of Canada's annual fish catch of more than 2×10^9 lb (valued at over $200 million) could be substantially increased by the use of irradiation equipment.

The Mobile Demonstration Unit (Fig. 2) developed in the potato irradiation program has a throughput of 2000 lb/hr. Its ^{60}Co source delivers an 8000-rad dose to the potatoes with ±15% uniformity, the uniformity being achieved by irradiation of the potatoes on all four sides in a Ferris wheel type of carrier. The commercial plant built subsequently (Fig. 3) may be used for other foods and possibly some medical products. It has a vegetable storage unit with an initial capacity of 15 million pounds and a potential capacity of 45 million pounds. It uses the highly efficient "pallet-box" irradiation scheme (Fig. 4), in which the items are irradiated from two or more sides for

Fig. 2 Mobile ^{60}Co irradiator.

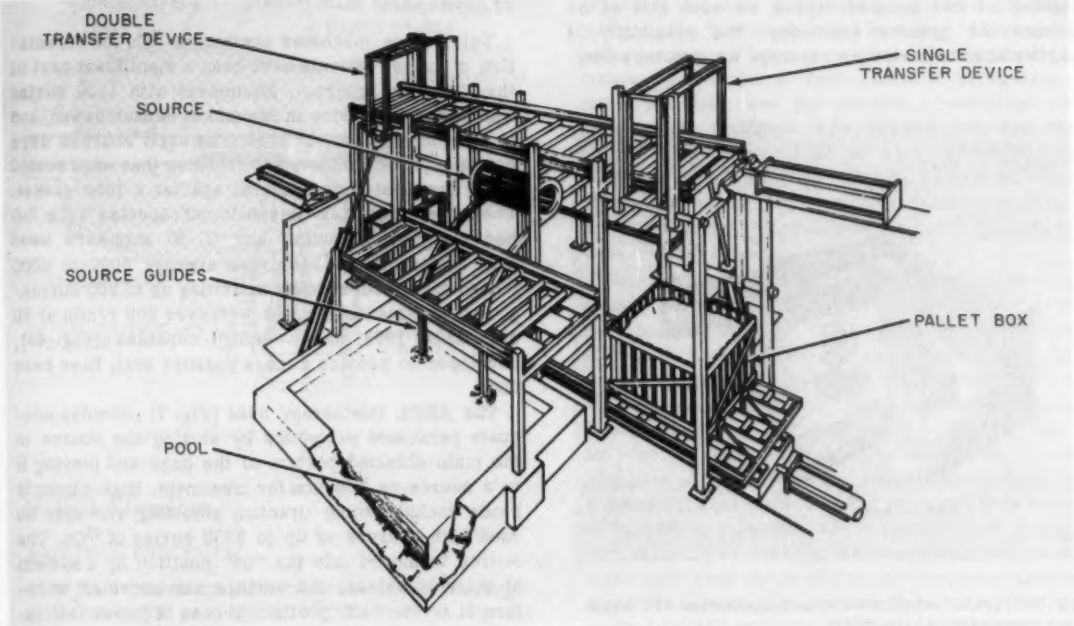

Fig. 3 Continuous-flow pallet-box industrial irradiator. The maximum throughput is 40 boxes per hour.

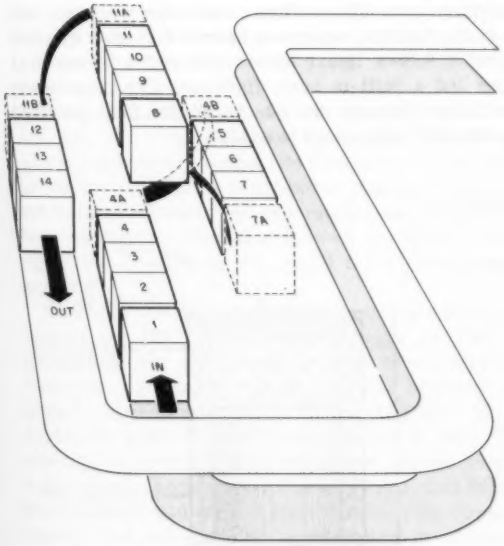

Fig. 4 Flow pattern of pallet-box irradiator.

more uniform dose distribution. Whenever possible, foods are irradiated in the usual storage and shipping container and without disturbance or tumbling of the product during the irradiation process.

PHARMACEUTICAL PRODUCTS

The Canadian concept of a ^{60}Co unit for radiation sterilization of pharmaceuticals evolved independently and was brought to fruition when Ethicon, Inc.,* became interested in the use of ^{60}Co for sterilizing medical products. The first such unit was built in England. Subsequently, Ethicon placed a contract with AECL for design and construction of three cobalt irradiation plants, two to be installed in the United States. The first began operation in Somerville, N. J., in August 1964, and the second, of the same design but lower throughput, was started up at San Angelo, Tex., in November 1964. Both are fully automatic and require only intermittent attention. The ^{60}Co source is stored in a pool of water and moves in a vertical plane between storage and operation. The product containers also move in a vertical plane during irradiation. A third plant, also commissioned in November 1964, is a small batch irradiator (Fig. 5) in Peterborough, Canada. It is designed to handle a small volume at low cost, and the source is kept in dry storage.

An irradiator recently completed for the Tasman Vaccine Laboratory in New Zealand is similar to the Somerville plant except that it is designed for three

*The Ethicon complex will be described in more detail in *Isotopes and Radiation Technology*, 4(4).

instead of two product layers on each side of the source for greater efficiency. The possibility of earthquakes indicated pool storage for greater safety.

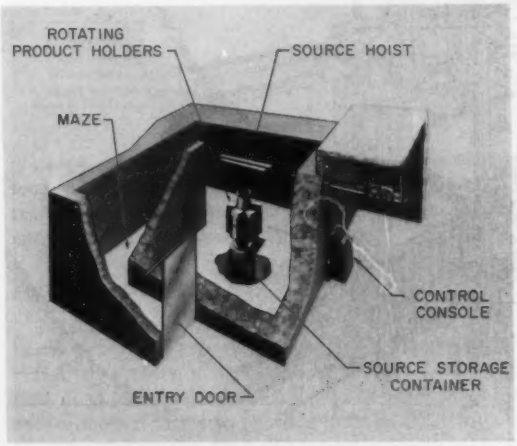

Fig. 5 Typical installation of a Gammabeam 850 batch-operation industrial irradiator.

CHEMICAL SYNTHESIS

A continuous-flow plant was designed and installed at the SAS Biodegradable Detergent Market Development Plant of the Esso Chemical Company at Bayonne, N. J. This plant is now producing sodium alkane sulfonate, an intermediate for Esso's biodegradable detergent.

TELETHERAPY

Teletherapy machines containing ^{60}Co for irradiation of cancer patients have been a significant part of the Canadian program. Equipment with 1100 curies was placed in service in Saskatoon, Saskatchewan, and in London, Ontario, in 1951. The early sources were contained in stainless-steel capsules that were sealed by a threaded cap tightened against a lead gasket. These were known as "international capsules" (Fig. 6*a*) because both Canadian and U. S. suppliers used this design. Today's sources average 3000 to 4000 curies each, with specific activities up to 200 curies/g, and provide dose rates well over 200 r/min at 80 cm. Since 1959, doubly welded capsules (Fig. 6*b*), developed to provide a more positive seal, have been used.

The AECL teletherapy head (Fig. 7) provides adequate personnel protection by storing the source in the main shielded portion of the head and moving it to a source-on position for treatment. High-capacity heads include some uranium shielding and may be used with sources of up to 9000 curies of ^{60}Co. The source is moved into the "on" position by a system of solenoid valves, and springs are provided to return it to the "off" position in case of power failure.

In this head the well-collimated beam with minimum penumbra essential for small-field curative therapy is obtained by use of a small source diameter and a cone-shaped multileaf diaphragm with telescoping trimmer bars. The isodose curve from a system with a 2-cm-diameter source compares favorably with that from a 4-Mev linear accelerator with a 5-mm focal spot and a built-in beam flattener. For large-scale palliative therapy, the 33- by 33-cm field provided in the AECL machines is used.

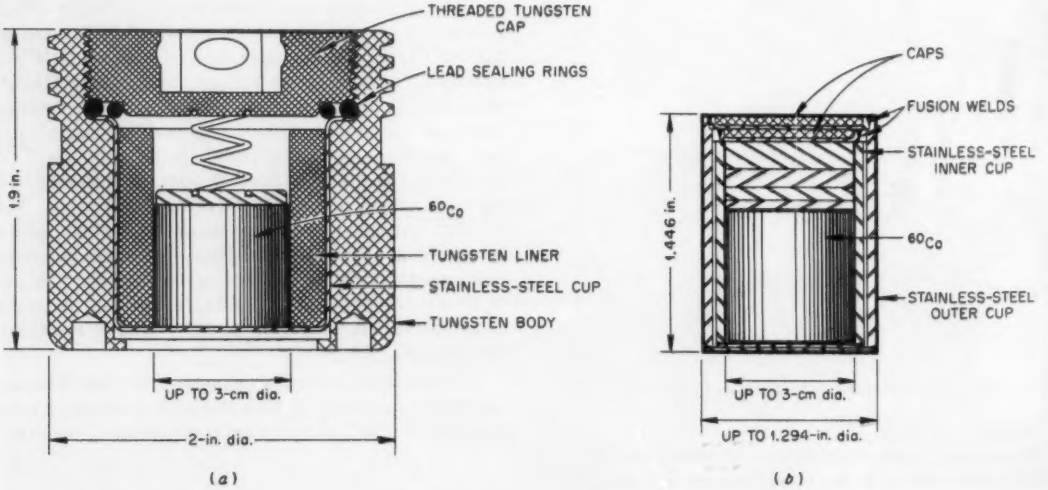

Fig. 6 Cobalt-60 teletherapy sources: (a) old international capsule; (b) present AECL capsule.

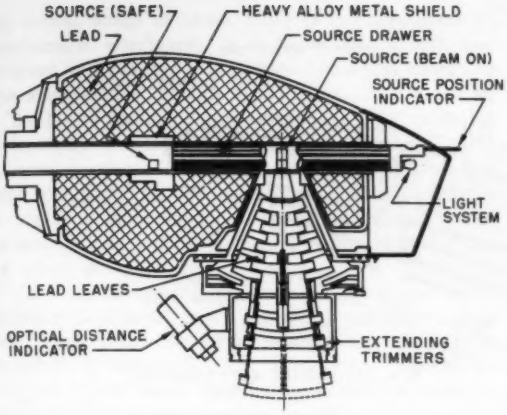

Fig. 7 AECL teletherapy head.

United Kingdom

RADIATION PROCESSING

Of the nuclides that have been considered for large-scale radiation processing—^{60}Co and ^{137}Cs—by far the most work has been done with ^{60}Co. Enough of this nuclide to meet demands for the next 5 years is expected to be produced at an acceptable cost by the UKAEA's Calder Hall and Chapel Cross reactors. If necessary, production can be expanded by using the civil power reactors of the Central Electricity Generating Board and possibly some of the prototype reactors.

There are no immediate plans for large-scale extraction of ^{137}Cs from fuel-processing raffinates. Large amounts are potentially available—10 Mc/year by 1970—from the nuclear power program, and both extraction processes and separation plant design have been extensively studied at Harwell and Risley. However, the demand is too small for recovery to be economic.

Research and development work on radiation processing in the United Kingdom began in 1955 at Harwell. It was transferred in 1956 to the Wantage Research Laboratory, which now has probably the largest concentration of irradiation facilities in the world: 23 experimental irradiation cells, with ^{60}Co sources of between 100 and 10,000 curies, plus a 4-Mev linear accelerator. The three principal industrial uses of radiation—sterilization, food preservation, and initiation or acceleration of chemical processes—have been studied. As yet, sterilization processes are the only ones that have been commercialized in the United Kingdom, but some promising applications in the other fields are appearing.

Medical Supplies Sterilization. In 1960 a pilot package irradiation plant (Fig. 8) began operation at Wantage. It has been outstandingly successful in achieving its aim of providing facilities for determining data that can be used in design, construction, and operation of large gamma-irradiation plants; for commercial firms to test radiation processing of their products; and for service irradiations until commercial facilities are adequate. It can treat standard packages (13.25 by 11.5 by 8.5 in.) with doses of from 0.070 to 10 Mrads. Its present source strength of about 300 kc of ^{60}Co enables the plant to irradiate about 300 cu ft of packages per day to a dose of 2.5 Mrads.

Data obtained at Wantage were used in the design of the four commercially owned plants (Table 1) that have been built in the United Kingdom for radiation sterilization of medical supplies. Their present total source strength is ~500 kc, and, together with the Wantage plant, they are annually irradiating about $45 million worth of products. A striking feature of the operation of these plants—reliability in service—is illustrated by the experience of Johnson's Ethical Plastics plant at Slough. In more than 4 years of operation, stoppages due to failures have averaged less than 1% of the total operating time, and most of these have been due to electrical-component failures. Regular scheduled maintenance has required only about 1 week's shutdown of the plant each year.

Wantage-developed data were also used in designing the Westminster Carpet Co. plant in Australia for sterilizing goat hair. With its present source strength of 750 kc of ^{60}Co, this is the largest commercial irradiation plant in the world. Two large plants for sterilizing medical supplies now being commissioned in West Germany also incorporate design features from Wantage experience.

In its capacity as a service irradiation facility, Wantage sterilizes practically all the medical dressings for the British Army. It also sterilizes plastic tubing, valves, catheters, bags, and such unusual items as ball-point pens for marking on patients in the operating theater and sandpaper for use by plastic surgeons in various hospitals. In commercial irradiation plants, the most numerous items sterilized are disposable hypodermic syringes and surgical sutures; some 160 million syringes and 40 million sutures have been gamma sterilized in the United Kingdom during the past 6 years.

Food Preservation. Wantage studies on food preservation by irradiation have shown, as they have elsewhere, that the dose used must be high enough to achieve the desired effect but low enough to avoid deterioration of taste and odor. Recent work on radiation pasteurization of fish to prolong shelf life is showing promising results. Following studies on elimination of *Salmonella* from foods by 0.5 to 1 Mrad of gamma irradiation, 10 tons of horsemeat was successfully processed in the frozen state. Specific-pathogen-free and completely sterile rations have been prepared for animal colonies in research laboratories.

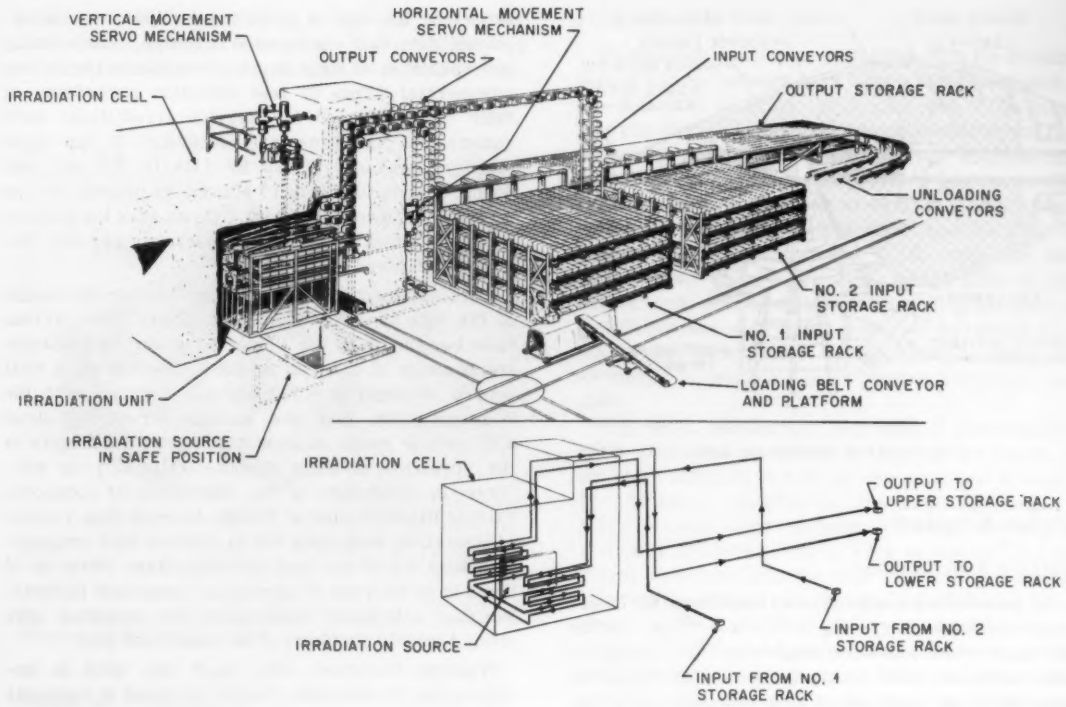

Fig. 8 Wantage package irradiation plant.

Table 1 COBALT-60 IRRADIATION PLANTS IN THE UNITED KINGDOM

Plant owner	Startup date	Source capacity, kc	Purpose
UKAEA	1960	500	Trial irradiation of industrial products
Johnson's Ethical Plastics Ltd.	1961	700*	Sterilization of disposable syringes
Ethicon Ltd.	1962	200	Sterilization of sutures
Gillette Industries	1964	750	Sterilization of disposable syringes
Swann-Morton Ltd.	1966	64	Sterilization of scalpel blades

*Given in Table 1 of Harmer article (page 267 of this issue) as 300 kc.

TELETHERAPY

The use of ^{60}Co for teletherapy in the United Kingdom began in 1954 with a kilocurie unit, presented by AECL, at the Mount Vernon Hospital. Today there are sixty-eight ^{60}Co units in U. K. hospitals with a total source strength of 170 kc. There are six ^{137}Cs units with a total of about 10 kc.

Some 70 kc of ^{60}Co per year is now being supplied in this field. However, the newer sources require a high specific activity—150 curies/g or more—and some of this will have to be imported for the present. The only U. K. reactors capable of producing such material are the DIDO and PLUTO research reactors at Harwell, and these cannot fill the whole demand. A newly designed control rod will produce some high-specific-activity ^{60}Co, but even this will not be enough.

HEAT SOURCES

A limited program on the development of radioisotope heat sources has been carried out in the United Kingdom in the last few years, with attention concentrated on possible commercial markets for radioisotope-powered generators. The most promising prospect appears to be for navigational lights where, with conventionally powered sources, maintenance and replacement costs are often considerable.

The first four flashing-light devices, the RIPPLE units, were developed and constructed to use 600 to 700 curies of ^{90}Sr as the titanate obtained from ORNL. They have an electrical output of less than 0.1 watt. The next stage of the program aims at development of three more-powerful units, which will be loaned to marine authorities for trials of their performance and suitability. Estimates of the possible future demand for generators of this type indicate that in the next 10 years the requirement may reach 1000 watts of electrical power, representing

an annual usage of about 3 Mc of ^{90}Sr (assuming 5% conversion efficiency). It is hoped that a sufficient market will develop that private firms will undertake the manufacture and sale of the units.

So far work has been limited to ^{90}Sr, which is potentially available in very large amounts as a fission product. However, extraction at a price that will make its use economic is possible only on a scale of some 10 Mc/year, and a demand of this magnitude has not yet developed. Work on production of other possible heat sources—^{238}Pu, ^{242}Cm, and ^{244}Cm—is proceeding at Harwell, but again, in the absence of a large-scale demand, there are no plans for large-scale production. However, the United Kingdom is definitely going ahead with ^{237}Np extraction, of which some 20 kg will probably be available from the civil power program by 1970.

commercialization stage, private industry requested the Japanese government to provide large-scale installations. Accordingly, in 1961, a central organization devoted to radiation chemistry research was set up within the Japan Atomic Energy Research Institute to coordinate private, university, and government research.

In 1964 the Takasaki Radiation Chemistry Research Establishment was set up as a branch of this Institute. Its first intermediate-scale experiment was polymerization of styrene with staple fibers that had been preirradiated with 2-Mev electrons. Next, they made polyethylene from ethylene gas, using ^{60}Co irradiation (Fig. 9). This product is now being evaluated, but work on the process is continuing. Radiation polymerization of trioxane, using the radiation from a 3-Mev Cockcroft-Walton accelerator, is about to

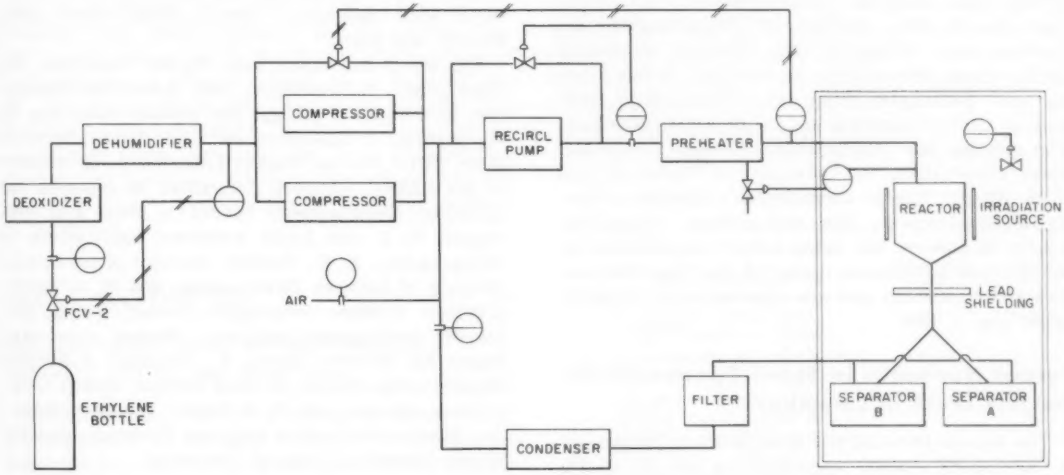

Fig. 9 Vapor-phase polymerization of ethylene by irradiation.

Japan

Japanese radiation chemistry research has been concentrated on producing and improving the properties of high-molecular-weight materials by means of irradiation. Early studies led to the establishment, in 1956, of the Japanese Association for Radiation Research on Polymers. When the work reached the

be launched on a 10-kg/batch scale. Radiation techniques that have been developed include introducing a radioactive gas into a reaction system to act as a source. Oxidation of benzene and chlorination of hydrocarbons in the presence of radiation are being studied, and experiments on the graft polymerization of fibers and radiation-induced chemical reactions in nuclear reactors are being done.

Section

VII

Isotopes and Radiation Technology

Miscellaneous

Conferences

Second Low-Energy Symposium Held at University of Texas

The Second Symposium on Low Energy X- and Gamma Sources and Applications was held at the Texas Union Building of the University of Texas, Mar. 27–29, 1967. Attended by 120 participants, the meeting was devoted to five sessions: Mössbauer Spectroscopy, Biomedical Applications, X-Ray Fluorescence Techniques, Progress in Radioisotope Analysis in Other Countries, and Industrial Applications. The meeting was jointly sponsored by the Texas Nuclear Corporation, the University of Texas, and the U. S. Atomic Energy Commission's Division of Isotopes Development. The proceedings, comprising nearly 50 papers, are being edited and published by the Isotopes Information Center of Oak Ridge National Laboratory (ORNL) and are expected to be available about Aug. 1, 1967.

Second Nucleonics in Space Symposium Set for July 12–14 in Columbus

The Second International Symposium on Nucleonics in Aerospace, jointly sponsored by the U. S. Air Force (Air Force Flight Dynamics Laboratory, Flight Control Division), the U. S. Atomic Energy Commission, and the Instrument Society of America, will be held at the Sheraton Columbus Hotel, Columbus, Ohio, July 12–14, 1967. Approximately 70 papers have been submitted, and the program will include sessions on Detectors, Nondestructive Testing, Sources (Low Level), Analytical Techniques, Gaging, Ranging and Guidance, Health and Safety, and Heat Sources. The registration fee for the 3-day session is $20, including the banquet. For further information, write R. C. Beavin, Control Elements Research Branch (FDCL), Air Force Dynamics Laboratory, Wright-Patterson Air Force Base, Ohio 45433.

UCLA Conference on Atomic Energy Attracts Scientists and Educators

The University of California at Los Angeles (UCLA) and the Inter-American Nuclear Energy Agency cohosted a conference March 22–28 that attracted nuclear leaders and educators from Latin America and the United States. The conference on Applications of Nuclear Energy in Latin American Development was planned to attract representatives of international and federal nuclear organizations, industry, educational institutions, and electric utilities to discuss their problems in the peaceful applications of nuclear energy. The six participating Latin American countries were Argentina, Brazil, Chile, Costa Rica, Mexico, and Peru.

The topics discussed were nuclear desalting, nuclear power, radioisotopes, inter-American cooperation and nuclear energy, and nuclear education. In the session on radioisotopes, T. E. Hicks, Director of the UCLA Nuclear Energy Laboratory and member of the USAEC Advisory Committee on Isotopes and Radiation Development, headed a panel that discussed U. S. and Latin American applications of radioisotopes. E. E. Fowler, Director of the USAEC Division of Isotopes Development, and P. S. Baker, Director, Isotopes Information Center, ORNL, discussed development programs. Papers were presented by Roberto Maass E., Director of Nuclear Medicine Department, National Nuclear Energy Commission, Mexico, and V. P. Guinn, Technical Director, Neutron Activation Analysis Program, General Atomic Division of General Dynamics.

IAEA Conferences Well Attended by U. S. Scientists

Thirteen conferences of the International Atomic Energy Agency (IAEA) have been or will be attended by U. S. nuclear scientists and engineers in 1967. The first two meetings were held in March. One was held at Monaco and covered radioactive dating and methods of low-level counting; the other was held in Brussels and treated the use of plutonium. Of the 11 remaining conferences, 5 will be in Vienna and 1 will be in the United States. Neutron Thermalization and Reactor Spectra will be discussed on July 17–21 at the University of Michigan, Ann Arbor; the USAEC will be cosponsor.

The 11 remaining conferences are: Symposium on Containment and Siting of Nuclear Power Plants, Vienna, April 3–7; Nuclear Activation Techniques in the Life Sciences, Amsterdam, May 8–13; Radiosterilization of Medical Products, Budapest, June 5–

9; Disposal of Radioactive Wastes into the Ground, Vienna, May 29–June 2; Instruments and Techniques for Assessment of Airborne Radioactivity, Vienna, July 3–7; Neutron Thermalization and Reactor Spectra, University of Michigan, Ann Arbor, July 17–21; Thermodynamics of Nuclear Materials, Vienna, September 4–8; Use of Isotope and Radiation Techniques in Soil Physics and Irrigation Studies, Istanbul, June 5–9; Use of Isotopes in Studies of Nitrogen Metabolism in Soil–Plant–Animal System, Sofia, Bulgaria, August 28–September 1; Symposium on Heavy Water Power Reactors, Vienna, September 11–15; Symposium on International Extrapolation & Comparison of Nuclear Power Costs, London, October 9–13.

Radioisotope Techniques Institute

The Sixth Annual Radioisotope Techniques Institute will be conducted from June 5–30, 1967, by the Western New York Nuclear Research Center. The 120 hr of instruction in the theory and handling of radioisotopes is divided into 60 hr of lectures and 60 hr of laboratory exercises using the Nuclear Research Center facilities. Further information and application forms are available from Radioisotope Techniques Institute, Western New York Nuclear Research Center, Incorporated, Power Drive, Buffalo, N. Y. 14214.

AEC Activities

AEC Increases Prices of Radioisotopes, Markets ^{123}I for the First Time

Effective April 21, AEC raised the prices of 15 radioisotopes. At the same time, ^{123}I will become available for sale, on a special order basis because of its short half-life (13 hr).

The new prices are: ^{122}Sb, $9/mc; ^{37}Ar, $45/mc; ^{131}Ba, $180/mc; ^{210}Bi. $30/mc; ^{72}Ga, $12/mc; ^{130}I, $3.80/mc; ^{109}Pd, $3/mc; ^{142}Pr, $9/mc; ^{186}Re, $6/mc; ^{153}Sm, $7.50/mc; ^{111}Ag, $15/mc; ^{187}W, $18/mc; ^{131}I, $1/mc; ^{170}Tm, $300/curie; and ^{133}Xe, $100/curie. Minimum orders for ^{170}Tm and ^{133}Xe are $150; all others are $25.

These radioisotopes have industrial, biomedical, and other research uses. Iodine-123 is especially useful in diagnosing thyroid conditions.

Details of the new price and delivery schedules are available from Oak Ridge National Laboratory, Isotopes Sales Department, Isotopes Development Center. P. O. Box X, Oak Ridge, Tenn. 37830.

AEC Withdraws from Production and Sale of ^{87m}Sr Generators

The medically important radioisotope, ^{87m}Sr, will no longer be available from the AEC as of April 30. It was available from AEC's Brookhaven National Laboratory in generator, or "cow," form.

The chief use of ^{87m}Sr is as a tracer in the study of bone fractures; it is one of a group of radioisotopes that are useful because of their short half-lives and low energies.

The policy of the AEC is to withdraw from production and sale of any radioisotope that private industry can and does provide; commercial demands for ^{87m}Sr generators are now being met by private organizations at reasonable prices.

AEC Publishes License Fees for Comment

Acting in line with established government policy, AEC has proposed that fees be charged for its licenses. This is legal under Title V of the 1952 Independent Offices Appropriation Act. General licenses would not require a fee; neither would licenses for export or import. Fees would be charged for construction and operation licenses for nuclear reactors as well as other production facilities, for specific licenses for uranium and thorium source materials, for radioisotopes, and for ^{233}U, ^{235}U, and plutonium. The notice was published in the *Federal Register* on Mar. 11, 1967. Table 1 lists the proposed fees.

Table 1 SCHEDULE OF MATERIALS LICENSE FEES*

Material category	Application fee	Annual fee
1. All material licensed other than in categories 2 through 5	None	$ 25
2. By-product material issued under 10 CFR Part 33 or Part 35.11(e)	$100	$ 50
3. By-product material of 100,000 curies or more in sealed sources used for irradiation of materials	$500	$100
4. Special nuclear material in quantities sufficient to form a critical mass	$300	$100
5. Waste disposal (receipt of waste by-product material, source material, or special nuclear material for commercial disposal by land or sea burial)	$500	$200

*If a license falls within more than one category, the license shall be deemed to fall within the category for which the fee is highest.

Reactor-Licensing Organization Streamlined to Stress Safety

Harold L. Price, AEC's Director of Regulation, has announced changes in the organization of the regulatory staff aimed at streamlining its reactor-licensing functions and increasing emphasis on the development of safety criteria and standards. The growing number of applications to construct and operate nuclear power plants was cited as one reason for the reorganization.

The increased emphasis on safety and standards is reflected in the formation of a new Division of Reactor Standards and the reorganization of the Division of Safety Standards as the Division of Radiation Protection Standards. The functions of the old division have been divided between the two new divisions. Edson G. Case will be Director of the Division of Reactor Standards, and Forrest Western, formerly Director of the Division of Safety Standards, will be Director of the Division of Radiation Protection Standards.

The Division of Reactor Standards will develop and recommend safety standards, criteria, and guides for the location, design, construction, and operation of reactors; it will also develop and recommend rules applicable to facility licenses, provide technical advice to the other AEC divisions on reactor safety, coordinate the regulatory staff's participation in the AEC's reactor-safety research and development program, and maintain liaison with other organizations.

The Division of Reactor Licensing has established three groups to handle the different aspects of its work: a reactor projects group, a technology group, and a reactor operations group. The enforcement function, formerly the responsibility of the Division of State and Licensee Relations, has been given to the Division of Compliance. The new organization is shown in Fig. 1.

AEC Releases Four New Films

Four new nontechnical films are available from the AEC. "Atoms on the Move: The Transportation of Radioactive Materials" covers the different methods of transporting radioactive materials; "Miracle in the Desert: The Story of Hanford" is a documentary of the AEC's Hanford facility. "Farm Fresh to You" treats the application of radiation to fruits and vegetables to increase shelf life, and "The Fresher the Better" presents information on radiation preservation of seafood. Both films on food are about 13.5 min long; the other two films last about 25 min. All four films are in color.

"The Fresher the Better" gives a simple definition of radiation and by cartoons shows how radiation affects the spoilage organisms in seafoods. Scenes show fish being processed in a seafood packing house, then being transported to the Marine Products Development Irradiator where they are irradiated by ^{60}Co gamma rays. The great number of taste evaluations and chemical and physical tests to determine any differences between irradiated and nonirradiated seafoods are shown. Results are given of a comparison of 3-week-old irradiated fish to unirradiated fish less than 1 day old; the irradiated fish tasted just as good to the testing panel. Research at Louisiana State University, Washington State University, and Michigan State University is mentioned. The film points out that the treatment extends shelf life of refrigerated fish by a factor of 3 or 4, and the creation of a fresh-fish market in the Middle West is stressed.

"Farm Fresh to You" also starts with a brief popular-level definition of radiation and its effect on spoilage organisms. The film describes the research efforts at the University of California at Davis and at the University of Florida to stop the great economic loss caused by food spoilage. Although food spoilage is more a problem for industry than for the housewife, she will benefit greatly from a solution to the problem since she will have a greater variety of foods throughout the year to serve her family. A brief rundown is given of the different fruits and vegetables being studied. A large portion of the film deals with strawberries, which are considered the most promising fruit for marketing after irradiation. Potatoes, wheat and wheat products, and bacon are named as irradiated foods already cleared by the Food and Drug Administration (FDA) for consumption. As in the film on seafoods, taste-panel evaluations and research into chemical and physical similarities between irradiated and unirradiated foods point out that the only difference between irradiated and nonirradiated fruits and vegetables is that the irradiated foods keep longer.

In "Atoms on the Move," the differences between various radioactive materials are presented as are the potential hazards involved in their shipping. The film shows typical shipments enroute by train, truck, aircraft, and ship. The responsibilities of such agencies as the AEC, Interstate Commerce Commission, Bureau of Explosives, Federal Aviation Agency, Coast Guard, and state authorities are discussed. Safety research and development are emphasized.

"Miracle in the Desert" traces the history of the Hanford Engineering Works from the discovery of plutonium in 1941 to construction in 1943 under wartime secrecy with postwar additions and improvements up through the New Production Reactor—the first reactor to produce plutonium for military needs and to produce steam to generate electricity. Hanford's broad programs in research into the peaceful uses of atomic energy are described.

Information on either loan or sale of the films is available from the Audio-Visual Branch, Division of Public Information, U. S. Atomic Energy Commission, Washington, D. C. 20545.

AEC Finds Radiation-Control Programs of 15 Agreement States Compatible

The Atomic Energy Commission has examined the radiation-control programs in each of the 15 states that have taken over part of its licensing and regulatory authority and has announced that the state programs continue to be both compatible with the Commission's program and adequate to protect health and safety.

* A/D = Assistant Director

Fig. 1 The AEC regulatory organization.

Alabama, Arkansas, California, Florida, Kansas, Kentucky, Mississippi, Nebraska, New Hampshire, New York, North Carolina, Oregon, Tennessee, Texas, and Washington are responsible for regulating within their borders the uses of radioisotopes, small quantities of fissionable materials, and uranium and thorium.

The information on which the evaluation was based came from the continuing exchange of data with the states, from semiannual meetings with each state at which the state's program is reviewed, and from annual meetings with all agreement states. The Commission also examined the status of the states' regulations, the qualifications of their regulatory personnel, and the states' procedures for inspection, licensing, and enforcement. This is the second formal redetermination by the Commission.

Arizona and Louisiana Become 16th and 17th Agreement States

The states of Arizona and Louisiana have assumed responsibility for licensing, rule making, and enforcement of the uses of radioisotopes, source materials (uranium and thorium), and small quantities of fissionable materials within their borders. An agreement was signed in Phoenix, Ariz., on Mar. 30, 1967, by Governor Jack Williams and Chairman Glenn T. Seaborg of the U. S. Atomic Energy Commission. On April 17 Wilfrid Johnson of the USAEC and Governor John McKeithen signed an agreement at Baton Rouge, La. Louisiana's agreement became effective May 1, Arizona's on May 15.

The Louisiana Board of Nuclear Energy, Division of Radiation Control, will carry out that state's radiation-control program. There are 214 AEC licenses in Louisiana. The responsible Arizona agency will be the Arizona Atomic Energy Commission. There are about 100 AEC licenses in Arizona for the use of radioactive materials.

Use of ^{46}Sc Exempted by AEC

AEC regulation 10 CFR Part 30 is being amended to exempt from licensing the possession and use of ^{46}Sc when incorporated in a synthetic plastic resin. The manufacture of the resin will still require a license, however.

Chevron Research Co., Richmond, Calif., plans to make the scandium-containing resin. The resin will be injected into an oil well and polymerized in place to consolidate the sand and prevent its entering the oil, plugging the well, and limiting oil production.

AEC Exempts Tritium and Promethium and Increases Tritium Limit

The AEC is amending its regulations to permit a greater quantity of tritium to be used in luminous aircraft signs. The use of tritium in luminous marine navigational aids and the use of ^{147}Pm in spark-gap tubes are to be exempted from licensing.

United States Radium Corp. asked that the amount of tritium in its aircraft safety signs be increased from 4 curies to 10 curies. The brightness of the signs had to be increased to meet requirements of the Federal Aviation Agency; the increase in tritium content will also give the signs a longer useful life. The signs are safety devices that operate even when the aircraft electrical system fails.

Marine compasses and navigational instruments for measuring boat speed, water depth, and wind direction, which use tritium to illuminate the dials and pointers of the instruments, can now be owned without a license from the AEC. The petition by Brookes and Gatehouse, Inc., asked that the limit be set at 750 mc of tritium for the compasses and 250 mc for the navigational aids.

Siemens America, Inc., successfully petitioned for the exemption from licensing of the use of ^{147}Pm in spark-gap tubes (surge-voltage protectors). The tubes protect signaling systems, communications systems, open-wire lines and cables, and other electrical equipment against sudden overvoltages that might damage the equipment. The ^{147}Pm ionizes the rare gas used in the device and assures a fast reaction time of the tubes; up to 30 μc may be used.

The manufacture or import of the spark-gap tubes would still require a specific license from the AEC.

Isochem and Five Others Compete for Hanford Separations Contract

The Atomic Energy Commission has announced that the contract for operating the Hanford chemical separations plant—the "200" area—is open because of Isochem, Inc.'s, decision not to build the Fission Products Conversion and Encapsulation (FPCE) plant. Isochem has operated the plant, which recovers plutonium and uranium from nuclear waste materials, since Dec. 31, 1965; but the contract was contingent upon Isochem's constructing the FPCE. In January, however, Isochem decided that increased costs of construction and failure of the market for the separated fission products to develop meant that Isochem could not go ahead with construction plans as it had intended.

When Isochem and AEC negotiated the termination of the FPCE construction contract, the second half of the contract—operation of the chemical separations plant—became open. The facilities operate at an annual cost of about $24 million, employ about 1400 people, and represent an investment by AEC of about $270 million. In addition to demonstrating ability to manage such a large technical operation, proposers must demonstrate a willingness to promote further commercial ventures in the Richland–Kennewick–Pasco area. This is part of the AEC policy of co-

operating with industrial development efforts of communities where AEC plants are large employers.

Originally, expressions of interest in the chemical-separations plant contract came from 10 companies. Those still competing are Aerojet-General Corporation, Allied Chemical Corporation, Dow Chemical Co., TRW Systems, Atlantic Richfield Co., and Isochem itself. Other companies are expected to bid.

Proposals must be in by May 15, and AEC tentatively plans to start negotiations by June 15.

ORNL in 1966 Doubles 1965 Total for Curies of Radioisotopes Sold

Oak Ridge National Laboratory sold 2.611 million curies of radioisotopes in 1966. Although this more than doubled the 1965 previous total of 1.281 million curies sold, sales were mainly of ^{60}Co, ^{137}Cs, and ^{90}Sr. The total of 5322 shipments in 1966 was below the number of shipments in 1965, a decline that has been continuous for the past several years. Private industry has assumed production and distribution of routinely prepared radioisotopes, and ORNL is doing more research and development. The new total does reflect the fact that orders are becoming bigger and bigger. During the 20 years since the first radioisotope shipment in 1946, ORNL has made more than 177,000 shipments representing nearly 6.9 million curies of radioisotopes.

Almost 15,000 Licenses Issued for Radioactive Materials

As of Dec. 31, 1966, 14,702 licenses had been issued for use and possession of radioactive materials, according to the USAEC's Division of State and Licensee Relations. The two largest categories of license holders were (1) medical institutes and physicians and (2) industrial firms.

The 15 agreement states at the end of 1966 had issued 5387 licenses to 4384 licensees. Medical institutions and physicians had 2182 licenses issued to 1709 licensees, and there were 1721 licenses granted to 1545 industrial firms.

AEC itself issued 8313 By-product Material licenses to 5998 licensees, with 2847 licenses going to 2076 medical institutions and physicians and 2970 licenses going to 2207 industrial firms. In addition, AEC issued 1002 Source and Special Nuclear Material licenses.

Availability of Isotopes and Services

ORNL Stable-Isotope Inventory Augmented

Recent additions to the Oak Ridge National Laboratory enriched stable-isotope inventory include:

Isotope	Purity, %	Amount available, g	Isotope	Purity, %	Amount available, g
^{24}Mg	99.9	63.0	^{108}Cd	81.4	1.9
^{25}Mg	97.9	22.0	^{111}Cd	91.3	8.0
^{26}Mg	99.7	50.0	^{112}Cd	95.7	15.6
^{60}Ni	99.9	200	^{144}Nd	97.5	36.4
^{64}Ni	97.9	5.1	^{154}Gd	66.9	12.8
^{63}Cu	99.7	25.8	^{178}Hf	94.7	14.8
^{67}Zn	89.5	28.3	^{179}Hf	87.0	10.0
^{70}Zn	79.6	1.6	^{196}Hg	40.1	0.6
^{109}Ag	99.1	9.0	^{204}Hg	92.6	2.8

ORNL Offers Radiometric Analysis by Gamma-Ray Spectrometry

Quantitative radiometric analysis of samples by nondestructive gamma-ray spectrometry is now available from the Analytical Chemistry Division of Oak Ridge National Laboratory. When the spectrometer output can be resolved by one of several computer codes now being used, the cost of analysis is a small fraction of the cost of analysis by other methods. As many as 15 radionuclides have been determined quantitatively in a single sample by this method. For further information, write M. T. Kelley, Director, Analytical Chemistry Division, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37830.

Rad-Mat, Inc., Announces Irradiation Services

A new industrial radiation processing service center has opened for business at Gaithersburg, Md. Rad-Mat, Inc., uses a 3-Mev accelerator, producing both electrons and X rays for radiation processing, housed in a modern building with associated support equipment and 10,000 sq ft of warehouse space. Special regulating systems guarantee good dose reproducibility. The electron radiation is 3 Mev at 10 ma; the X-ray radiation is 10,000 r/min. The conveyor system can handle objects up to 4 ft by 4 ft by 4 ft. Current prices are listed in Table 2; a special price list applies to irradiation of polyethylene tubing, coated wire, and sheets. For further information, contact Rad-Mat, Inc., P. O. Box 485, Gaithersburg, Md. 20760.

Table 2 CUSTOM IRRADIATION PRICE SCHEDULE

Time	Charge, $
First 15 min	75
Second 15 min	64
Third 15 min	56
Fourth 15 min	50
Each additional 15 min	50

Neutron Products Announces Prices of ^{60}Co from Big Rock Point

Singly encapsulated ^{60}Co gamma sources are now available from Neutron Products, Inc. The sources will usually be furnished as hollow sources, i.e., the cobalt is in a wound-spring form; but Neutron Products reserves the right to provide solid sources if they desire. If the solid source is provided, the activity will be 10% above that requested. Hollow sources come in tubes 12 and 10 in. long; solid sources are 12, 10, and 5.375 in. long.

The ^{60}Co is provided in two grades—standard and premium. The standard has an average activity of 1000 curies per source; the premium has an average activity of 1500 curies per source. Prices are given in Table 3. Discounts are given for orders placed more than 60 days before scheduled shutdown of the Big Rock Point reactor where the ^{60}Co is produced.

Table 3 PRICES OF NEUTRON PRODUCTS, INC. ^{60}Co*

Quantity, curies	Standard grade, U. S. $/curie	Premium grade, U. S. $/curie
50,000–75,000	0.65	0.70
75,000–100,000	0.60	0.65
100,000–250,000	0.55	0.60
>250,000	0.50	0.55

*F. O. B. customer's plant in customer's cask. Does not include shipping, insurance during shipment, and $200 cask-loading charge.

For further information, write Neutron Products, Inc., 1730 M Street, N. W., Washington, D. C. 20036.

General

Prostate Uptake of ^{69m}Zn

A "first" in the experimental, medical use of radionuclides was achieved recently at Walter Reed General Hospital. The prostate gland was scanned with a scintillation detector using 14-hr ^{69m}Zn (0.44-Mev gamma). This is the first known successful scan performed with ^{69m}Zn on the prostate in humans. The scan reveals areas of tissue that have taken up radioactive material, and such identification of the prostate could lead to an effective method for early detection of prostatic carcinoma.

One of the patients selected for this study was scheduled for a prostatectomy. This allowed the medical researchers to perform an in vivo scan before surgery and an in vitro scan of the gland itself after surgery. Figure 2 is a composite picture showing posterior and anterior in vivo and in vitro scans with the actual prostate in the center.

It has been demonstrated that the prostate gland will concentrate injected zinc; however, as yet there is no evidence that malignant tissue will accumulate zinc in a significantly different manner from normal prostatic tissue. If such a difference in zinc uptake does exist, the technique will be a valuable tool for early detection of prostatic carcinoma.

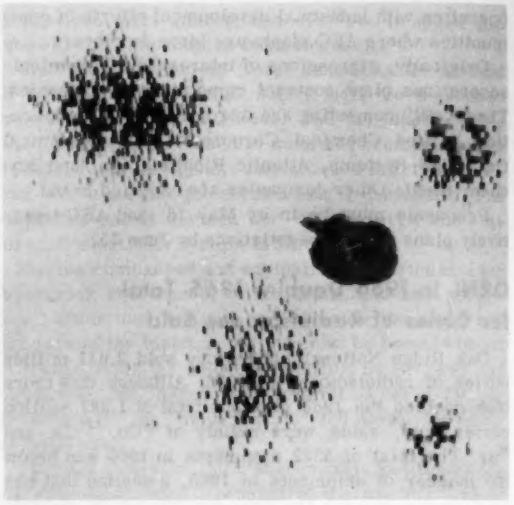

Fig. 2 A composite of prostate gland scans demonstrating ^{69m}Zn activity. The scans on the left of the subject's prostate are in vivo scans taken 1 day after injection of the isotope; the scans on the right were taken after surgery 2 days after injection. Anterior scans are above and posterior scans below the prostate gland in the figure.

Medical researchers from Walter Reed General Hospital and The James Buchanan Brady Urological Institute of The Johns Hopkins Hospital developed the prostate scanning procedure on dogs and monkeys using 243-day ^{65}Zn (1.1-Mev gamma).* The relatively long half-life and high radiation energy of ^{65}Zn are undesirable for human scanning; the short half-life of ^{69m}Zn and its lower radiation energy are much better suited for this purpose.

The ^{69m}Zn was prepared at the Isotopes Development Center of ORNL by the neutron irradiation of highly enriched ^{68}Zn in the Oak Ridge Research Reactor and supplied to Walter Reed Hospital as part of a cooperative development program. (H. B. Hupf)

Food-Irradiation Expert Accuses Industry of Foot Dragging

The food industry has been apathetic toward irradiation of food, charged Dr. Walter M. Urbain at the National Canners Association meeting in Chicago, Jan. 22–25, 1967. Despite the fact that 20 years of research has been done on food irradiation, there are no commercial food-irradiation plants built or

*G. S. Johnston, R. P. Gibbons, J. D. Schmidt, G. P. Murphy, and W. W. Scott, Fluid and Tissue Kinetics of ^{65}Zn in the Dog and Monkey, *Investigative Urology*, 3: 419–426 (1966).

planned. While admitting that there are technical problems remaining to be solved, Dr. Urbain insisted that this method of food preservation has more than enough promise to stimulate active industry interest.

Dr. Urbain, a Michigan State University professor with more than 20 years experience in the food industry, put most of the blame for the situation on the FDA. He claimed that the FDA should ask outside counsel—possibly from the National Academy of Sciences—to review its requirements for approval of food irradiation; he termed those requirements "tough" and "controversial." In the same length of time that the FDA has taken to clear 3 foods, the Surgeon General's Office has cleared 22.

Urbain also stated that the cost of irradiation is assumed to be high, but that no one knows for sure. He estimated that radiopasteurization could cost as little as 0.3 cent/lb.

Industry could help accelerate acceptance of the method, which Urbain says damages food no more than the conventional heat methods do, by supplying efficacy data to FDA and by settling the cost question. If industry markets it, the consumer will also accept irradiated food easily.

FDA Labeling Order on Irradiated Food Definitely Includes "Radiation"

The Food and Drug Administration, in an order effective 30 days after it was published Mar. 2, 1967, has stated that foods treated with radiation of any kind must have the word *radiation* on a label affixed to the food. The kind of radiation does not have to be named; the word *ionizing* can be substituted for *gamma, X-ray,* or *electron.* Food is *treated* with low doses and *processed* with high doses. In addition, bulk shipments of food treated with low doses of radiation must be labeled *do not irradiate again.* So far, no definite decision has been made on AEC's suggestion that the benefits of radiation be identified on the label, i.e., "Treated with radiation *to retard spoilage*" or "Treated with radiation *to extend shelf life.*"

Hazleton Labs Publishes Up-to-Date FDA Petition List

Hazleton Laboratories, Inc., is keeping track of all petitions to FDA related to irradiation of food. This status list will be updated every 6 months, according to E. A. Garlock, Senior Consultant. Table 4 shows those petitions and their status as of Feb. 10, 1967.

Iso Nuclear To Bid on Meat Irradiator

Financial support by the Midland Capital Corp., a New York investment firm, has enabled Iso Nuclear to bid on the pilot-plant meat irradiator to be built for the AEC. They would build at the Western New York Nuclear Service Center and lease land from the New York State Atomic and Space Development Authority; the proposed plant would have a capacity of 2×10^6 lb of meat per year. Iso Nuclear would also work on development of commercial meat products with John Morrell & Co.

Neptunium Recovery Plant Construction Recommended for West Valley, N. Y.

A report submitted to the New York State Atomic and Space Development Authority (ASDA) by an independent consultant recommends that a plant for the recovery of neptunium be constructed adjacent to the Nuclear Fuel Services (NFS) facility at West Valley, N. Y. for startup by 1969. The plant would be a Multi-Purpose Plant designed for recovery from NFS waste of Np, Am, Cm, Cs, Sr, Pm, and Ce; but market considerations led to the conclusion that only the neptunium part should be constructed now.

Total cost of the Multi-Purpose Plant was estimated at $10.7 million, with 5 plants—each recovering a specific radioactive product—costing $8 million and support facilities costing $2.7 million. The neptunium recovery facility would cost $1.49 million. The four other recovery plants were designed to be added on individually at any time after the construction of the support facilities and the neptunium recovery plant. The Multi-Purpose Plant design was based on handling the waste stream from NFS's processing of 500 tons per year of fuel with a burnup of 25,000 Mwd/ton. This would be about 143 thousand liters.

Operating costs for the plants are estimated at $3.7 million per year, averaged over 7 years, with the neptunium plant costing $759,000. Product recovery costs of the neptunium are expected to average $27/g over 7 years; encapsulation costs are expected to average $10/g. Costs of other isotopes are given in Table 5.

The report concludes that a plant of the type described is capable of producing the isotopes at a cost competitive with other sources. Further, the demand for neptunium is such that construction for startup in 1969 is recommended. A decision by ASDA on whether and how to proceed with the project will be made later this year.

FDA Petitioned for Use of ^{210}Po

The 3M Company has filed a petition with the FDA asking for a regulation on the use of ^{210}Po as a source of alpha-particle radiation for the elimination of static electricity in food-packaging operations. The purpose of the petition is to discover whether or not FDA considers such a use of polonium a food additive. If it is not so considered by FDA, it will be outside FDA's jurisdiction.

Table 4 REGULATIONS AND PETITIONS ON RADIATION PRESERVATION OF FOODS, FEB. 10, 1967

Product	Petitioner	Source	Dose	FDA petition			FDA regulation			Remarks
				Filing date	Ref., Vol.	F.R.* Page	Issue date	Ref., Vol.	F.R.* Page	
Bacon	U. S. Army	^{60}Co	4.5–5.6 Mrads	8-17-62	28	8214	2-8-63	28	1465	-
Bacon	General Electric Company	5-Mev electrons	4.5–5.6 Mrads	6-5-63	28	5537	8-30-63	28	9526	-
Bacon	U. S. Army	10-Mev electrons	4.5–5.6 Mrads	8-23-63	28	9329	4-21-65	30	5631	-
Bacon	AEC	^{137}Cs	4.5–5.6 Mrads	12-18-63	28	13797	4-1-64	29	4672	-
Bacon	Radiation Dynamics, Inc.	X rays from 5-Mev electrons	4.5–5.6 Mrads	7-23-64	29	9910	12-19-64	29	18056	-
Wheat, wheat products	Brownell et al.	22-Mev gammas	20,000–50,000 rads	10-4-62	28	1465	8-21-63	28	9208	-
Wheat, wheat products	High Voltage Engineering Corp.	5-Mev electrons	20,000–50,000 rads	12-18-63	28	13797	2-26-66	31	3196	-
Wheat, wheat products	AEC	^{137}Cs	20,000–50,000 rads	9-1-64	29	12481	10-10-64	29	14027	-
White potato	AEC	^{137}Cs	5,000–10,000 rads	4-1-64	29	12481	10-10-64	29	14027	-
White potato	U. S. Army	^{60}Co, ^{137}Cs	5,000–10,000 rads	1-30-65	30	1013	10-10-64	29	14027	-
White potato	U. S. Army	^{60}Co	5,000–10,000 rads	6-6-63	28	5588	7-8-64	29	9329	-
White potato	U. S. Army	^{60}Co, ^{137}Cs	15,000 rads	1-30-65	30	1013	11-9-65	30	11801	-
Citrus (oranges, lemons, and grapefruit)	AEC/U. S. Army	^{60}Co, ^{137}Cs	75,000–500,000 rads	12-26-63	28	13797	-	-	-	Pending; see amendment
Citrus (oranges)	AEC/U. S. Army	^{60}Co, ^{137}Cs	75,000–300,000 rads	-	-	-	-	-	-	Amendment to Citrus; submitted for filing 5-4-65
Marine products (haddock, pollock, ocean perch, cod, flounder, and sole)	AEC/U. S. Army	^{60}Co, ^{137}Cs, 10-Mev electrons, 5-Mev X rays	100,000–200,000 rads	9-15-65	30	11801	-	-	-	Pending
Strawberries	AEC	^{60}Co, ^{137}Cs	100,000–250,000 rads	5-18-66	31	7256	-	-	-	Pending
Ham	U. S. Army	^{60}Co, ^{137}Cs	3.5–5.6 Mrads	8-24-66	31	11241	-	-	-	Pending
Packaging materials, nine classes of materials	AEC	Gamma	1 Mrad	2-8-64	29	2318	8-14-64	29	11651	-
Packaging materials amendments										
Cellophane (Saran coated)	AEC	Gamma	1 Mrad	2-18-65	30	9116	6-11-65	30	7599	-
Nylon	AEC	Gamma	1 Mrad	9-8-65	30	11400	-	-	-	-
Polyester (Saran coated)	AEC	Gamma	1 Mrad	7-30-65	30	9551	-	-	-	Pending
Polypropylene (Saran coated)	AEC	Gamma	1 Mrad	7-30-65	30	9551	-	-	-	Pending
Six films	U. S. Army	Gamma	6 Mrads	7-21-65	30	9116	-	-	-	Pending
Parchment paper	U. S. Army	Gamma	6 Mrads	1-15-65	30	547	3-12-65	30	3354	-

*F.R. = *Federal Register*.

Table 5 ESTIMATED PRODUCTION COSTS AT THE MULTI-PURPOSE PLANT*

Isotope	1st year cost, $	7th year cost, $	Average cost, $
^{137}Cs (encapsulated)	0.37/curie	0.05/curie	0.10/curie
^{90}Sr (encapsulated)	0.39/curie	0.06/curie	0.11/curie
^{147}Pm (unencapsulated)	0.037/curie	0.006/curie	0.011/curie
^{241}Am (unencapsulated)	234.0/g	31.0/g	58.0/g
^{244}Cm (unencapsulated)	468.0/g	81.0/g	148.0/g

*Assuming construction concurrently with the neptunium plant and operation to recover these isotopes to the extent they are available.

Availability of New-Design Electromagnetic Separator Announced by Colutron Corporation

A new design, flexible, lightweight isotope separator is available from the Colutron Corporation, Boulder, Colo. The basic difference between the Colutron, which is named for the University of Colorado where it was developed, and the older sector-type machine is the analyzing system; the Colutron uses a velocity filter instead of a sector magnet. Some of the Colutron's chief advantages in modern research stem from this difference. Since the analyzing system weighs about 500 lb instead of 10 or 15 tons, the Colutron is much smaller and lighter than other isotope separators; it can be moved more easily.

Because of the velocity filter, the Colutron has adjustable mass dispersion. This means that the separator can collect any number of isotopes simultaneously—all the isotopes of any element or all the elements and compounds from M = 1 to M = 500 in one operation. Another feature that improves the operation is a vacuum lock for rapid transfer of collected isotopes from the deceleration chamber. This is necessary to prevent oxidation when thick targets are deposited on thin backing.

The Colutron is said to be simple and reliable and does not require extra technical personnel. More detailed information, including price and delivery data, is available from the Colutron Corporation, P. O. Box 1288, Boulder Colo. 80301.

IAEA Passes Million-Dollar Mark in Medical Isotope Research

The International Atomic Energy Agency (IAEA) has now spent more than a million dollars to aid work with radioisotopes in medicine, principally to make techniques available to developing countries. Out of 84 contracts, 13 went to Africa, 15 to the Middle East, 16 to Latin America, and 17 to Southeast Asia and the Far East. Table 6 gives the distribution of funds by subject.

Table 6 DISTRIBUTION OF IAEA FUNDS FOR MEDICAL RADIOISOTOPE RESEARCH

Subject	Number of contracts	Cost, $(U. S.)
Bone metabolism	14	94,400
Anemia	23	272,625
Endemic goiter	18	318,265
Malnutrition	16	220,510
Parasitology in man	11	175,550
Miscellaneous	2	8,683
	Total	1,090,033

University of Florida Announces Radiological Health Specialist Program

Financial support for a limited number of qualified students interested in radiological health is now available from the Department of Bioenvironmental Engineering, University of Florida, Gainesville, as part of the expanded program in bioenvironmental engineering. Candidates for either the M.S. or Ph. D. degree will receive a broad base in traditional sanitary engineering and may specialize according to individual interest from courses in the Departments of Chemistry, Physics, Nuclear Engineering, Radiology, and Bioenvironmental Engineering.

The Department of Bioenvironmental Engineering will occupy a new four-story building in July 1967, containing about 5000 sq ft of floor space. Its laboratories are fully equipped with up-to-date radiation-detection instrumentation.

For further information, write Department of Bioenvironmental Engineering, University of Florida, Gainesville, Fla. 32601.

ORNL Isotopes Division and USDA Develop Radioisotope Eggshell Gage

R. S. Pressly of the Isotopes Division of Oak Ridge National Laboratory, in cooperation with the U. S. Department of Agriculture, has developed a pinpoint-size source of radioactive material that can be used to measure the thickness of very thin objects. The U. S. Department of Agriculture is using the source in a new device that tests the strength of eggshells. There is a direct correlation between thickness and strength of eggshells.

The source is a minute quantity of $^{106}Ru/^{106}Rh$ set into the point of a pencil-like rod. Other possible uses of the device are the gaging of crustacean-shell thickness, nutshell thickness, or even coatings of the outer surface of an object. With slight modifications it could determine the quantity of bone present in commercial foods such as chicken salad or hamburger.

Radiation Curing of Coatings—Economics and Competitive Claims

Kennard Morganstern, president of Radiation Dynamics, Inc., presented data at a meeting of the National Association of Coil Coaters in October 1966 which compared radiation curing of coatings with conventional oven curing. He based his comparison on the curing of a 2-ft-wide strip at 150 ft/min and claimed the following: investment was $80,000 for a Dynacote accelerator vs. $100,000 for an oven, paint system costs were $0.70/lb for electron curing vs. $1.10/lb for oven curing, and operating costs were $5/hr for the accelerator vs. $9/hr for the oven. Other advantages claimed were that the accelerator only takes up 10 ft of linear floor space while the oven requires 100 ft. Also, the accelerator is ready to go within 5 min; the oven is not ready for 2 hr after startup.

At the 16th Congress of the Society of Automotive Engineers in January, Morganstern predicted that the automotive industry would greatly increase the use of radiation to cure coatings. He stated that, in addition to greater speed, the accelerator could cure acrylic monomer coating at $0.25/sq ft vs. the $0.65/sq ft required for a conventional oven. The operating cost of the radiation-curing system is less than the $0.40/sq ft difference.

Meanwhile, Ford Motor Co.—active in the radiation-curing field already—is claiming that its 300-kev accelerator now operating at Boise, Idaho, costs only one-third to one-fifth as much to operate as those of its two chief competitors, Radiation Dynamics and High Voltage Engineering Corp.

Electron Accelerator Used To Cure Building Products at Two New Plants

At Plainfield, Ill., a 500-kev accelerator (insulating core transformer) from High Voltage Engineering Corp. is being used by Radiation Polymer Corp. to process prepainted building materials. At Boise, Idaho, two accelerators with Ford Motor Co.'s patented modification [see *Isotopes and Radiation Technology*, 4(1): 73 (Fall 1966)] are being used to cure coating on wood paneling.

Company officials of Radiation Polymer Corp., which was organized in mid-1965, claim that the cost of curing the building materials with radiation is 8 to 25% below conventional methods. The company

intends to treat papers, textiles, and plastics and to provide technical skills and assistance to other firms.

Oil Consumption of New Buick Engines Measured with ^{82}Br Before Installation

Engineers with General Motors Research Laboratories are using ^{82}Br to check any Buick engine for large use of oil before it is installed in a new car. The program began in the fall of 1966, but it was only announced at the first of this year. Buick checked five different engine makes in the early stages of development; a new test facility at the production-line plant in Flint, Mich., is being expanded to check about 1% of the 2500 engines built there each day. The 80 mc of ^{82}Br used each week are prepared at the University of Michigan, Ann Arbor.

AECL To Upgrade Newfield Irradiator for Irradiation Experiments

The Newfield Products Ltd. potato irradiator will be used by Atomic Energy of Canada Ltd. as a research facility. The irradiator was leased from AECL, and, when the potato irradiation facility failed because the potato crop was the worst in 35 years, the irradiator and its cobalt was returned to AECL. The ^{60}Co source will be strengthened from its present 27,000 curies to 100,000 curies. The package-handling mechanism, which was designed for potatoes, will be modified to treat other fruits and vegetables as well. AECL may also use the facility for some commercial irradiation work. This work will be coordinated by the Commercial Products Group at AECL in cooperation with the Quebec government.

Martin Marietta Corp. Expands Line of Radioisotope Electric Generators

Martin Marietta Corp. of Baltimore has more than doubled its line of radioisotope-fueled electric generators. The units were first offered for commercial sale in the spring of 1966; eight 25-watt units were listed as off-the-shelf items [see *Isotopes and Radiation Technology*, 4(1): 74 (Fall 1966)]. Now models at two more power levels and lightweight models in aluminum are being added to the list.

The two new power levels are 50 watts and 3 watts. The LCG (Low Cost Generator)-50 comes in A, B, and C models. The first two are standard terrestrial and subsea models; the LCG-50C is a lightweight aluminum model, intended for terrestrial use. Like the LCG-25 units, the LCG-50 electric generators are fueled with ^{90}Sr in titanate form; they are offered at a fixed price, have an AEC-approved design, are capable of being refueled easily, and have a guaranteed power output. The LCG-50 is 48 in. high and 30 in. in diameter; the LCG-25 is 35 in. high and the same diameter as the LCG-50.

The MW-3000 is a 3-watt model intended for deep-sea use. It weighs 3000 lb, has a 5-year power-output warranty, and can operate at depths of 15,000 ft. It is 24 in. in diameter and 30 in. high. One MW-3000 has been sold to the Navy.

Aluminum models of the LCG's are being offered for sale, at costs of about 20% more than the standard models. The lightweight LCG-25 weighs only one-third as much as the 3000-lb standard LCG-25; the weight of the LCG-50 is reduced from 4800 lb to 1700 lb by the use of aluminum.

Prices of the Martin energy sources range from $20,500 for the MW-3000 to $109,400 for the LCG-50 in lots of 10 each.

Radioactive Dirt, 20 Years Old, Brings $880,000 at Auction

Just after World War II, the Atomic Energy Commission stockpiled uranium ore at various places in the United States. Some was stored near St. Louis. This was later processed by a St. Louis firm for the AEC; however, much low-grade ore was left. A chemist reported there was Cu, Ni, Co, and Se in the ore as well as U. In 1966 the low-grade ore—essentially dirt—was bought by Continental Mining and Milling Company who intended to process it for the minerals it contained—the value is now estimated at $7.5 million. However, the firm ran into financial difficulty. When its St. Louis holdings were auctioned in February this year, the radioactive dirt was purchased by the Commercial Discount Corporation for $880,000. About 100,000 tons of dirt now await chemical processing—if anyone can decide how to go about it economically. No one seems to be able to do so.

New England Nuclear Adds ^{87m}Sr Cow to Isotope-Generator Line

New England Nuclear Corporation (NEN) on Mar. 23, 1967, announced that it was adding an ^{87m}Sr isotope generator to its line of products. The announcement was followed shortly by the AEC's announcement that it would withdraw its ^{87m}Sr generator from sale. NEN is now offering four "cows" for sale; the other three are ^{99m}Tc, ^{113m}In, and ^{68}Ga.

The new generator is prepared from enriched stable ^{87}Sr, purchased from Oak Ridge National Laboratory and irradiated in ORNL's 86-in. cyclotron. The resulting ^{87}Y is sorbed onto a resin bed in a glass column. Strontium-87*m* is removed by eluting with 5 to 10 ml of dilute $NaHCO_3$. The cow comes in two models, containing 5 mc and 10 mc of ^{87m}Sr, respectively. NEN's data sheet says that more than 25 mc of ^{87m}Sr can be milked from the 5-mc source over a 10-day period.

New England Nuclear's ^{99m}Tc generator is available in sizes of 100 mc or 200 mc of ^{99}Mo. Over 100 mc of ^{99m}Tc per week can be milked from the smaller

generator. The ^{113m}In generator comes in four sizes, with the largest size—charged with 25 mc of ^{113}Sn—yielding about 2500 mc of indium over 6 months. A ^{68}Ga cow is available in three sizes, ranging from 100 μc to 2 mc of ^{68}Ge.

New England Nuclear Corporation recently merged with Pilot Chemicals, Inc. Pilot makes plastic scintillators for radiation detection, organic fluorescent chemicals for liquid scintillation counting, and other products. Pilot will operate as a corporate subsidiary of NEN, and Pilot chairman, M. Hyman, Jr., will join NEN's board.

Use of Radioisotopes in the Electrical Industry

A 1-day conference* on the use of radioisotopes in the electrical industry was sponsored by the Institut für Rationalisierung und Organisation der Elektroindustrie (IRO) in Dresden, East Germany, in September 1965. Frau Prof. Dr.-Ing. L. Herforth, Director of the Institute for the Use of Radioactive Isotopes of the Dresden Technical University, served as chairman.

Seven papers were presented at the conference. The first paper was a review of 145 references covering tracer use, radiography, and gaging applications. It was followed by a paper listing 23 possible applications of radioisotopes in the East German electrical industry, which included wear measurements in numerous pieces of equipment, thickness and level gaging, and radiography. A paper on the use of radioactive tracers for studying wear of copper–graphite brushes included a review of 67 references. One paper discussed radioisotope uses in the specialized field of electronics, e.g., for studying deposition of luminescent material in television picture tubes, transistor-housing leak testing, and defect distribution in semiconductors. Other papers presented work on continuous measurement of thickness and moisture in ceramic materials by means of radioisotope gages; studies on determination of the water content of raw materials and molds at the VEB Elektroporzellan (electro-porcelain) plant; and a listing of items of information useful for firms interested in introducing isotope techniques. (Martha Gerrard)

Radioactive Tracers Aid Recovery of Ammunition

Picatinny Arsenal has recently developed a technique† for locating recoverable ammunition items after they are proof tested in the field. Ammunition must be recovered to eliminate explosive hazards to personnel, to discover the reason for failure, or to preserve security. However, finding the duds is often quite difficult, especially in rough terrain.

Four radioisotopes were considered: ^{59}Fe, ^{124}Sb, ^{192}Ir, and ^{46}Sc. Scandium-46 was chosen because it has two high-energy gammas for detection, can be produced easily, and is safer than the other radioisotopes since it has the lowest energy beta emission of the four.

The ^{46}Sc is provided as a small piece of metal foil sandwiched between two layers of metallized Mylar. The label thus formed is attached to the ammunition by a pressure-sensitive adhesive film. However, under field conditions, fresh liquid adhesive was also applied to the ammunition to ensure attachment. With a 1-mc label, the ammunition could be located from 54 ft if the item was on the surface; if the item was buried under 3 in. of rocky soil, the maximum detection distance was 19 ft.

The Army has set up requirements, based on results of the field tests, for a special detector that is within the capabilities of the nuclear instrumentation industry. After a prototype has been built and tested, detectors are not expected to cost over $1000 each.

(F. E. McKinney)

ORINS 1967 Training Courses in Medicine, Research, and Activation Analysis

Courses on the use of radioisotopes in research and in medical diagnosis, as well as a special course in activation analysis, are being offered at the Oak Ridge Institute of Nuclear Studies. The research and medical courses last 4 weeks and cost from $25 to $100 per week, depending on the nature of the participant's sponsoring organization. The special course on activation analysis lasts 3 weeks.

The course on medical diagnosis gives physicians basic training and clinical experience in the application of radioisotopes to diagnostic procedures. The course will be given September 11 to October 6. The last 2 weeks of the course consist of clinical practice and will be limited to 16 participants. For anyone desiring only the basic portion of the course, an additional 3-week course is being offered November 27 to December 15.

The lecture section of the research course will cover radiation detection, instrumentation, nuclear theory, radiological safety, interaction of radiation with matter, and principles and practice of many radioisotope applications. Counters, detectors, spectrometers, dosimetry, activation analysis, isotope dilution techniques, radiochemical separations, and biological applications are covered in the laboratory demonstrations. This course is offered August 7 to September 1 and September 11 to October 6.

A special course in neutron activation analysis is scheduled for May 31 to June 17. Lecture subjects and laboratory demonstrations will cover the char-

*Proceedings published (in German) in 1966 as brochure No. 4 of the IRO, Karl Marx Strasse, am Flughafen, Dresden 808.

†S. Helf and J. W. McCahill, Recovery of Ammunition Items Using a Radiotracer Technique, Technical Report 3489, Picatinny Arsenal, February 1967.

acteristics of radiation, interaction of radiation with matter, and detection of radiation. Detectors will be discussed, especially scintillation counters and solid-state detectors; the theory of neutron activation analysis will be covered, and radiochemical separation procedures related to activation analysis will be discussed.

Union Carbide—Neisler Offer Isotope Generators

Union Carbide Corporation is now offering isotope generators for sale through its Sterling Forest Research Center and Neisler Laboratories, a subsidiary. A standard ^{99m}Tc "cow" is available, as well as a new sterile ^{99m}Tc cow; at the first of this year an improved ^{113m}In generator was offered.

The ^{113m}In generator uses 118-day ^{113}Sn; the indium is obtained by eluting with dilute hydrochloric acid solution. The cow yields > 90% of the theoretically available 1.7-hr ^{113m}In, with radiochemical purity > 99.98%. Sizes from 5 mc to 100 mc are available. Although sterile, pyrogen-free reagents are used to prepare the generator, the product is not pharmaceutically refined. If sterility is required, the product should be autoclaved or passed through a millipore filter.

A quick and effective test developed by Union Carbide to check for tin breakthrough uses the sensitive color reaction between tin in acid solution and hematoxylin. Absence of color shows that the eluate contains less than 0.1 ppm tin.

The molybdenum-free ^{99m}Tc cow is available in high chemical purity as a Neimotec generator in 100-, 150-, and 200-mc sizes. Generators are shipped every Friday afternoon, calibrated for Monday morning.

A new sterile ^{99m}Tc cow will be offered in late June in 100-, 200-, and 300-mc sizes. The generator is an alumina column loaded with ^{99}Mo, sealed with rubber septa, and sterilized by autoclaving. Sodium chloride is added under aseptic condition to elute sterile solutions of the pertechnetate ion, with elution accomplished by the use of evacuated bottles.

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†Single copies of these booklets may be obtained free from USAEC, P. C. Box 62, Oak Ridge, Tenn. 37830.

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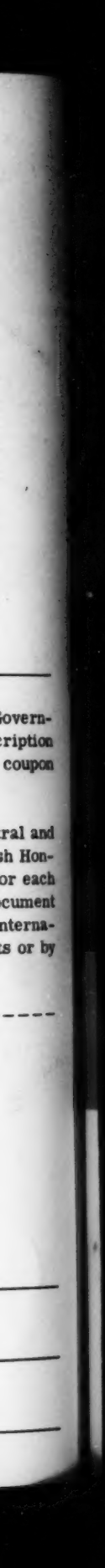
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